FINE ($PM_{2.5}$) AND COARSE ($PM_{2.5-10}$) PARTICULATE MATTER ON A HEAVILY TRAFFICKED LONDON HIGHWAY: SOURCES AND PROCESSES

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ABSTRACT

A large dataset for $PM_{2.5}$ and PM_{coarse} ($PM_{2.5-10}$) concentrations monitored near a busy London highway (Marylebone Road) has been analysed in order to define the factors that lead to high concentrations. The following have been highlighted as major influencing parameters: wind speed, prevailing wind direction (because of its role on the microscale dispersion within the street), the daily cycle of the atmospheric boundary layer (stable during the night/convective and mixed during the day) and traffic density. The average $PM_{2.5}$ and PM_{coarse} particle mass concentrations measured at Marylebone Road show daily and weekly cycles which correlate well with the traffic counts for heavy duty vehicles. The mainly diesel heavy duty vehicles are the main source of fine particulate matter at Marylebone Road. In particular lorries dominate PM_{10} exhaust emissions (median contribution of 42%) which are mainly in the fine (< 2.5 µm) size range. A strong correlation with PM_{coarse} suggests that the substantial turbulence induced by the heavy-duty traffic is responsible for the resuspension of particles from the road and/or that heavy-duty vehicles are larger emitters of dusts via abrasion processes. Median enhancements of approximately 7 µg m^{-3} for $PM_{2.5}$ (33% of $PM_{2.5}$) and about 4.5 µg m^{-3} for PM_{coarse} (45% of PM_{coarse}) due to the local traffic are found for Marylebone Road in comparison to the urban background of Bloomsbury. They correspond to a median heavy-duty traffic and light-duty traffic of about 290 vehicles per hour and about 3900 vehicles per hour respectively. At least 79% of the increment in PM_{coarse} concentrations is estimated to arise from non-exhaust vehicle emissions, which implies that at least 31% of PM_{10} vehicle emissions are non-exhaust emissions. An analysis of factors leading to high hourly concentrations of PM_{10} at Marylebone Road is conducted and reveals that these events were due either to building works or associated with periods of maximum heavy-duty traffic and favourable prevailing wind conditions. Lower wind speeds favour high $PM_{2.5}$ concentrations and stronger wind speeds favour high PM_{coarse} concentrations. Some episodes of high concentrations were associated with long range transport.

Key Words

PM_{10}, $PM_{2.5}$, non-exhaust vehicle emissions, light duty and heavy-duty traffic, resuspension, street canyon, fugitive dusts, long range transport

INTRODUCTION

The current European air quality standard for particle mass relates to PM_{10} and as a consequence the monitoring of airborne particulate matter in European urban areas is mainly based on the PM_{10} particle fraction. Particles below 10 µm correspond to the inhalable fraction of particulate matter and have been related to various adverse health effects. PM_{10} particle mass includes both fine (below 2.5 µm) and coarse (between 2.5 and 10 µm) fractions of airborne particulate matter which normally arise from different sources. Fine particles are mainly particles from combustion processes or are formed within the atmosphere by chemical processes. On the other hand, coarse particles generally arise from natural sources (wind-blown soil, sea spray, pollens) or from construction and quarrying activities. The regulation of PM_{10} particle mass requires a better knowledge of both $PM_{2.5}$ and PM_{coarse} sources, and their behaviour in the atmosphere. Pollutant dispersion mechanisms, depending both on the site topography and on the meteorological conditions strongly influence the concentrations of particles.

Transport, and more especially road transport is one of the most important sources of particulate matter, and is considered to be involved in all major air pollution issues including human health, ozone production, climate change, and acid deposition [1]. In London, where emissions from industrial sources and stationary combustion are modest, the traffic contribution has been estimated to represent 80% of the emissions of particles in 2001, while road transport corresponded to 19% of PM_{10} emissions for the entire United Kingdom [2].

Particle emissions from transport include contributions from exhaust emissions (both fuel and lubricating oil combustion), emissions from abrasion processes (tyre wear emissions, brake linings, catalyst deterioration etc) and road dust resuspension induced by the vehicle-generated turbulence. Road transport may also be responsible for a large part of secondary particulate matter formed via gas-to-particle conversion [3]. Whereas emissions of particles from engine exhaust may be estimated from dynamometer studies, non-exhaust particles are much more difficult to quantify, and measurements in the roadside atmosphere are a useful means of estimating the contribution of road transport to particulate matter concentration.

Particulate emissions from on-road vehicles contain a wide range of particle sizes from ultrafine particulate matter to coarse particulate matter. In this paper we focus on the fine (< 2.5 µm) and on the coarse (2.5 to 10 µm) fractions of particulate matter. The ultrafine fraction has been examined elsewhere [4].

This study is based upon atmospheric measurements on a busy road in London. Its purpose is to investigate (1) the contribution of light and the heavy-duty traffic to particle mass for $PM_{2.5}$ and PM_{coarse}, (2) the influence of meteorology and dispersion processes on the concentrations measured, and (3) the role of these factors and other sources in the occurrence of events of high concentrations of PM_{10}.

EXPERIMENTAL SECTION

SAMPLING SITE AND INSTRUMENTS

Sampling Sites

The Marylebone Road supersite is a part of the London Air Quality Network and is operated by the Environmental Research Group of Kings College, London. It is located on the kerbside of a major arterial route within the City of Westminster in London. The surrounding buildings form an asymmetric street canyon. Traffic flows of over 80,000 vehicles per day pass the site on 6 lanes with frequent congestion.

Particle data from the urban background of Bloomsbury in London are also used in this study. The Bloomsbury site is located in Russell Square Gardens in the centre of London at about 2 km from Marylebone Road. The gardens are laid to grass with many mature trees. Russell Square is surrounded by a 2/4 lane one-way road system (35,000 vehicles per day). The nearest road is at a distance of approximately 35 metres from the instruments.

Data and Instruments

Two Tapered Element Oscillating Microbalances (TEOM) are used to monitor PM_{10} and $PM_{2.5}$ particle mass concentrations at each site. The two TEOMs are identical instruments (Model 1400AB) except for the design of the sampling heads. The particle mass is determined by continuous weighing of particles deposited onto a filter. The filter is attached to a vibrating hollow tapered glass tube whose frequency changes as the mass loading on the filter increases. The vibration frequency is converted to mass concentrations by a microprocessor. Air at 16.67 L min^{-1} is sampled through the PM_{10} impactor inlet and divided between the filter flow (3 L min^{-1}) and an auxiliary flow (13.67 L min^{-1}). For $PM_{2.5}$ measurements, the TEOM is fitted with a URG $PM_{2.5}$ cyclone inlet. The inlet of the TEOMs is heated to 50°C prior to particles being deposited onto the filter in order to eliminate the effect of condensation or evaporation of particle water. This heating of the aerosol stream induces losses of semi-volatile species. A calibration factor has been established through an intercomparison of data from TEOMs and filter-based reference instruments in order to achieve the US EPA certification (1.03 *'TEOM reading' + 3 µg [5]. Despite this calibration factor, the TEOM instrument gives lower readings than filter-based methods in different sites in the UK as well as in other European countries [6, 7, 8, 9]. A previous study involving collocated Kleinfiltergerat (KFG) and Partisol samplers for the sampling of daily PM_{10} and $PM_{2.5}$ particle mass concentrations at Marylebone Road has revealed a mean KFG/TEOM ratio of 1.17 (Standard deviation of 0.30) for PM_{10} and mean Partisol/TEOM ratio of 1.10 (Standard deviation of 0.24) for $PM_{2.5}$. Correlations between daily particle mass from TEOMs and from gravimetric filter-based methods at Marylebone Road are not high. Ratios of gravimetric data/TEOM vary from one day to another. Differences between TEOMs and filter-based methods depend on the temperature, the relative humidity and on the concentrations of semi-volatile particulate compounds such as ammonium nitrate [10] and hence the use of basic correction factors is not adequate. A non-negligible part of particulate matter, probably semi-volatile compounds, is lost in the inlet of the two TEOMs and it should be borne in mind that the results and the interpretation are for virtually non-volatile particulate matter (to be precise, for particulate matter involatile at 50°C). From the PM_{10} and $PM_{2.5}$ measurements, the PM_{coarse} fraction corresponding to the mass of particles between 2.5 µm and 10 µm has been calculated by difference. The two sites are also equipped with continuous monitors measuring trace gases, including CO and NO_x concentrations.

Traffic counters have been installed at Marylebone Road since July 1998. All six lanes provide vehicle counts for 6 vehicle classes and 11 speed bins using a loop monitoring system. The loop monitoring system is based on the magnetic induction from the metal content of passing vehicles. The system at Marylebone Road has two loops in each lane allowing speed and vehicle classification according to the EUR6 scheme, based on the nature of the signal derived from the two sensors. In practice, vehicles are classified according to the length of their chassis. From the 6 classes available, we have formed two classes: the light duty class (cars and motorcycles) and the heavy-duty class (rigid lorries, articulated lorries, buses/coaches). During the period studied, Marylebone Road had an average traffic flow of 3600 vehicles per hour (ranging from 340 to 6035 vehicles per hour) with a mix of light duty vehicles (mean 90.5%, range from 77.6 to 99%) and heavy-duty vehicles (mean 9.5%, range from 1 to 22.4%).

Meteorological data from London wc Samos station (wind speed, wind direction, temperature, dew point) and from London Weather Centre station (rain data) and backwards trajectories were supplied by the British Atmospheric Data Centre. The data included in the present study are hourly data measured from July 1998 to July 2000 (2 years comprising 14,742 $PM_{2.5}$ and 14,413 PM_{coarse} measurements at Marylebone Road, i.e. more than 80% of possible data). Some 12,945 $PM_{2.5}$ and PM_{coarse} concentrations correspond to simultaneous measurements at Marylebone Road and Bloomsbury sites associated with meteorological data and traffic counts.

Emission factors from the U.K. National Atmospheric Emissions Inventory (NAEI) have been used to model the exhaust PM_{10} emissions from the mixed light duty and heavy-duty fleet at Marylebone Road. Average emission factors for the vehicle classes counted at Marylebone Road are determined using fleet-weighted PM_{10} emission factors for average speeds on UK urban roads; the composition of UK vehicle fleet technology (euro standard) and national proportions of diesel vehicles in cars and light good vehicles (LGVs) classes; emission scaling factors due to better fuels and technologies. The composition of the UK vehicle fleet is expressed as a fraction of vehicle km and takes account of decreasing annual mileage with increasing age of vehicle. In the absence of specific figures for Marylebone Road, national proportions of diesel engines in cars (9.9% in 1998; 10.9% in 1999; 11.5% in 2000) and in LGVs (7.4% in 1998; 8.1% in 1999; 8.4% in 2000) for urban roads are used in the calculations. The scaling factors take account of new improvements that reduce emissions from existing vehicles such as fuels of improved quality (e.g.

lower sulphur content of petrol and diesel fuels), vehicles retrofitted with emission abatement devices (e.g. particulate trap or oxidation catalyst fitted on diesel vehicles). Table 1 presents the average emission factors for the vehicle classes counted at Marylebone Road.

RESULTS AND DISCUSSION

Concentrations Measured and Influencing Parameters

Meteorological variables such as wind speed, wind direction, and traffic intensity are parameters that considerably influence the particle mass concentrations for Marylebone Road. Consequently, an examination of the influence of these parameters is conducted in this paper.

As shown in Figure 1, hourly concentrations of $PM_{2.5}$ and PM_{coarse} measured at Marylebone Road from July 1998 to August 2001 were highly variable. They ranged from 1 to 300 µg m^{-3} for $PM_{2.5}$ and 1 to 595 µg m^{-3} for PM_{coarse} and followed lognormal distributions (see on Figure 1, the fairly normal shapes of the distributions when a logarithmic scale is used for the X axes). Very high particle mass concentrations in comparison to most other sites in the UK are measured at Marylebone Road. More than 25% of hourly $PM_{2.5}$ concentrations were above 30 µg m^{-3} and more than 25% of hourly PM_{coarse} concentrations were above 15 µg m^{-3}. Some very high concentrations (above 100 µg m^{-3}) were occasionally measured for both fractions. No seasonal variation was observed for either $PM_{2.5}$ or PM_{coarse} at Marylebone Road. This suggests that there is no significant temporal variation of influential meteorological parameters and sources of particulate matter at Marylebone Road on a seasonal timescale, or that the effects of different factors cancel one another.

$PM_{2.5}$ and PM_{coarse} concentrations depend on the wind speed. Figures 2a and 2b present the median concentrations of $PM_{2.5}$ and PM_{coarse} measured for each 1-knot wind speed bin (1 knot = 0.515 m s^{-1}). The calculation of these medians smoothes the variability of concentrations due to the other influencing factors discussed below, especially the traffic flow. The median $PM_{2.5}$ concentrations decrease from about 25 µg m^{-3} to about 18 µg m^{-3} as wind speed strengthens from < 1 m s^{-1} to stronger winds > 9 m s^{-1}. This implies that dilution and dispersion due to the wind are responsible for a 30% reduction of $PM_{2.5}$ from weaker winds to stronger winds. A similar dependence on wind speed was found for $PM_{2.5}$ measured in the city of Birmingham [11]. In contrast, the coarse particulate matter concentrations increase when the wind speed increases. This pattern clearly reveals that at least a part of this particulate matter is from wind-driven resuspension processes. This wind-related resuspension process is considerable since the median concentrations for the weaker winds are about 11 µg m^{-3} and PM_{coarse} reaches concentrations close to 16 µg m^{-3} for stronger winds (that is to say an increase of 45%). The influence of the wind on coarse particles has been previously examined by Harrison et al. [12] who viewed the coarse particle fraction as comprising two components, a non-wind suspended component that corresponds to particles emitted from industrial and construction activities, traffic-induced resuspension and biological particles, and a wind-suspended component that arises mainly from natural sources such as sea spray and surface soils, or dusts on paved areas. The different behaviours of the two PM fractions with wind speed are in agreement with the commonly accepted view that most of fine particles arise from combustion processes or from gas-to-particle conversion within the atmosphere, while coarse particles arise mainly from mechanical processes.

The $PM_{2.5}$ and PM_{coarse} concentrations also vary strongly according to the prevailing wind direction (as measured above the canyon). The behaviour shown in Figure 3 at Marylebone Road is typical of street canyon with concentration roses very asymmetric towards the SSE direction, i.e. winds perpendicular to the direction of the road. A microscale meteorology is created by the surrounding buildings that determines the dispersion of the pollutants inside the Marylebone Road canyon. High concentrations (median concentrations of 25 µg m^{-3} for $PM_{2.5}$ and 13 µg m^{-3} for PM_{coarse}) are measured when the wind blows parallel to the street and a vortex situation is created when the wind blows perpendicular to the street. This re-circulation of air inside the street results in high concentrations of $PM_{2.5}$ (median of 22 µg m^{-3}) and PM_{coarse} (median of 13 µg m^{-3}) when the wind comes from a southerly direction (since the sampling site is located on the southern kerbside, measurements are influenced by on-road emissions). Much lower concentrations (median for $PM_{2.5}$: 13 µg m^{-3} ; median for PM_{coarse} : 7 µg m^{-3}) are observed when the wind blows from the opposite direction (measurements are less influenced by on-road emissions). This basic explanation considers Marylebone Road as a regular canyon. However, Marylebone Road forms an asymmetrical canyon (buildings of different height) with an intersection close to the sampling site. These explain the complex shape of the concentration roses in Figure 3. Pollutant dispersion in Marylebone Road has been described for carbon monoxide by Scaperdas and Colvile [13].

An influence of rain on $PM_{2.5}$ and PM_{coarse} concentrations is also observed. Both PM fractions decrease with heavy rain and increase during drought periods. However, this influence is weaker than the influence of the wind speed and wind direction and the results are scattered showing that other factors may influence the results (however a stronger relationship is found for PM_{coarse} particles). The closeness of the on-road fresh particulate matter source probably limits the impact of the washout effect.

The other parameter that has a significant influence on the concentrations measured at Marylebone Road is the traffic intensity. Figure 4 presents the median weekly cycles of $PM_{2.5}$ and PM_{coarse}. The median weekly cycles of the light duty traffic (cars and motorcycles) and of the heavy-duty traffic (lorries, buses, coaches) are also represented on this figure. Very similar patterns of traffic are observed from one week to another (except when there is a public holiday – such days are removed from the analysis) and the median weekly cycles of traffic densities well represent the single weekly cycles. The median weekly cycle of $PM_{2.5}$ and PM_{coarse} concentrations at Marylebone Road shows strong daily and weekly variations which correlate with those of the traffic. Higher concentrations are measured during daytime on workdays (median concentrations above 25 μg m^{-3} for $PM_{2.5}$ and above 15 μg m^{-3} for PM_{coarse}) and much lower concentrations during nighttimes and weekends. During the night, the median concentration of $PM_{2.5}$ is below 15 μg m^{-3} and of PM_{coarse} below 8 μg m^{-3}. The relation to traffic will be discussed further below.

The median $PM_{2.5}/PM_{10}$ ratio at Marylebone Road was about 0.67 for the whole period of study, i.e. on average two-thirds of the mass of the PM_{10} is within the $PM_{2.5}$ fraction. Interestingly, the same ratio was found for a busy road in Paris, France [14]and a similar one (0.76) for a road in Thessaloniki, Greece [15]. However, much lower ratios are found for some other kerbside sites located in the cities of Athens, Greece [16] and Bern, Switzerland [17]. These differences may be related to different meteorological conditions, traffic conditions or carriageway conditions. The ratio $PM_{2.5}/PM_{10}$ depends strongly on the wind speed as a result of the two effects, dilution of $PM_{2.5}$ and resuspension of coarse particles (Figure 2c). When the wind speed is low, the PM_{coarse} concentrations represent less than 30% of the PM_{10}

concentrations; while for stronger winds, the mass of PM_{10} is almost equally distributed between the $PM_{2.5}$ and the PM_{coarse} fractions. The other influential parameters have much smaller effects on the ratio $PM_{2.5}/PM_{10}$. APEG [8] reported that 90% of the mass of PM_{10} emitted from catalyst petrol and diesel exhaust, and 80% of the mass of PM_{10} emitted from non-catalyst petrol exhaust are in the below 2.5 µm size range. These ratios are much higher than the median ratio at Marylebone Road suggesting an excess of coarse particles in comparison to exhaust emissions.

Relation to Traffic

The weekly cycles of $PM_{2.5}$ and PM_{coarse} suggest that both PM fractions are related to the traffic. The highest concentrations are measured during the midday period of the workdays when the heavy-duty traffic is at a maximum level and reaches 20% of the total traffic flow. The lowest concentrations are measured on Sundays and a minor peak in concentration is seen on Saturday mornings. The $PM_{2.5}$ and PM_{coarse} cycles are clearly closer to the heavy-duty traffic cycles than to the light duty traffic cycle (see Figure 4). Pearson correlation coefficients have been computed from weekly cycles. Better correlations are found with the heavy-duty traffic (with $PM_{2.5}$ $r^2 = 0.60$; with PM_{coarse} $r^2 = 0.66$) than with the light duty traffic (with $PM_{2.5}$ $r^2 = 0.44$; with PM_{coarse} $r^2 = 0.41$).The stronger wind and air turbulence during the day would lead to the same PM_{coarse} concentration patterns on workdays as on weekends if the coarse particulate matter were only mechanically induced by the wind; this is clearly not the case.

The relationship between traffic data and particle data was also examined using a hierarchical cluster analysis (Wards' method and square Euclidean distance as a similarities measure). This multivariate method separates observations into groups according to their similarities and differences. The median weekly cycle of concentrations is used to smooth the variability of concentrations due to the variability of the meteorological parameters discussed above. Only the concentrations measured when the wind direction favours the measurement of on-road emissions are included in the analysis, that is to say the following wind directions have been considered: (1) wind parallel to the street (accumulation of on-road emissions), (2) and from southerly directions (vortex favouring the measurement of on-road emissions). The two tracers, the nitrogen oxides and carbon monoxide are simultaneously examined. Both are good tracers of on-road emissions. Carbon monoxide is mainly emitted by petrol vehicles, while nitrogen oxides are emitted both by petrol and diesel vehicles although the latter have higher emission factors than the former [8]. The results obtained with this method are presented on the dendrogram in Figure 5.

Two clusters are clearly formed:

- $PM_{2.5}$, PM_{coarse} and NO_x concentrations are clustered with the heavy-duty traffic
- CO concentrations are clustered with the light duty traffic

$PM_{2.5}$ and PM_{coarse} concentrations are associated with the heavy-duty traffic (lorries, coaches and buses). The clustering of nitrogen oxides with the heavy-duty traffic, and of carbon monoxide with the light duty traffic (mainly petrol vehicles) gives confidence in the results. The heavy-duty traffic is mainly comprised of diesel vehicles which have high emission factors for particle mass [18] and this result was expected for the fine fraction of particulate matter. The coarse particle mass was found to increase with wind speed suggesting that this fraction is comprised in part of resuspended material. The clustering of the coarse particulate matter with the heavy-duty traffic suggests that the substantial turbulence induced by the heavy-duty traffic is responsible for a greater magnitude of resuspension of particles from the road than the light duty traffic. Another explanation is the larger amount of dusts emitted by the heavy-duty traffic due to stronger abrasion processes including tyre wear and brake linings. Sternbeck et al. [19] found a strong correlation between particulate barium (a common element in brake linings not used as a fuel additive in Sweden) and heavy-duty traffic suggesting that heavy-duty vehicles are responsible for larger brake wear particulate emissions than light duty vehicles. Large vehicles have higher brake wear emission rates for airborne particles than small vehicles and these emissions would include both mechanically generated coarse particles and the formation of finer particles due to high temperatures at the brake/rotor interface [20]. Because of the substantial increment in coarse particle concentrations at Marylebone road in comparison to other sites in or close to London and the substantial workday to weekend difference, the influence of traffic on coarse particle concentrations measured at Marylebone road was expected [12]. Other studies have shown a link between coarse particulate matter and traffic flow [14, 17, 21] and chemical analyses of particulate matter from on-road vehicles have shown the impact of both road dusts [21, 22] and abrasion processes [19, 23, 24] on particulate matter. Harrison et al. [24] conclude that most trace elements in roadside aerosol are from vehicle wear products rather than exhaust emissions, which are dominated by carbonaceous compounds. This study indicates that heavy-duty vehicles are larger emitters of coarse particles from resuspension and/or abrasion processes than light duty vehicles.

Emission factors have been used to model the exhaust PM_{10} emissions from the mixed light duty and heavy-duty fleet at Marylebone Road. Results (Figure 6) show that PM_{10} exhaust emissions are dominated by heavy-duty vehicles and more especially by rigid lorries, while, vehicle counts are strongly dominated by light vehicles. On average, heavy-duty vehicles (8.6% of traffic) and lorries (7.5% of traffic) contribute 62% and 42% of PM_{10} exhaust emissions respectively (and up to 74% and 55% of PM_{10} exhaust emissions respectively). That is in agreement with the stronger correlations found between measured $PM_{2.5}$ and PM_{coarse} and the heavy-duty traffic. The weekly cycle of modelled PM_{10} exhaust emissions is very similar to the weekly pattern of $PM_{2.5}$ and PM_{coarse} concentrations measured at Marylebone Road. It shows that similar PM_{10} concentrations would be generated on weekends and workdays in the absence of heavy-duty traffic. The similarity of weekly variations in coarse particle concentrations with the weekly pattern of modelled PM_{10} exhaust emissions suggests that lorries also dominate non-exhaust vehicle emissions.

The concentrations measured near a road are assumed to be the sum of the urban background of the city and the enhancement due to local traffic. In contrast, the urban background is assumed to derive from a mixing of different sources (including on-road emissions, industrial activities etc) affected by different dilution factors.

A comparison between the particle mass concentrations measured at Marylebone Road and those measured at Bloomsbury (an urban background site in London) allows an estimation of the enhancement of particle mass at Marylebone Road due to the local traffic. Bloomsbury, although close to Marylebone Road, does not give a perfect indication of Marylebone Road’s urban background, and some higher concentrations of $PM_{2.5}$ and PM_{coarse} are occasionally measured in Bloomsbury. These higher concentrations result in negative enhancements for Marylebone Road that have no physical meaning. Much more negative enhancements (and very low values) are found for PM_{coarse} in comparison to $PM_{2.5}$

and negative enhancements are rarely simultaneous for $PM_{2.5}$ and PM_{coarse}. This implies that Bloomsbury is occasionally influenced by more or stronger sources than the urban background of Marylebone Road and that these sources are different for $PM_{2.5}$ and PM_{coarse}. Roadworks and construction activities locally contribute to very high concentrations of PM_{coarse} and may explain the occasional high concentrations measured in Bloomsbury. The less numerous higher concentrations of $PM_{2.5}$ are more difficult to explain. For this reason, only median enhancements are considered in this study.

Figure 7 shows that for both $PM_{2.5}$ and PM_{coarse} the median enhancement depends on the traffic intensity. It is at a minimum during the night and the weekend when the heavy-duty traffic is low (below 5 µg m^3 for $PM_{2.5}$ and below 3 µg m^3 for PM_{coarse}), and at a maximum during the midday period when the heavy-duty traffic is highest (above 12 µg m^3 for $PM_{2.5}$ and above 7 µg m^3 for PM_{coarse}). On the other hand, the weekly patterns of $PM_{2.5}$ and PM_{coarse} concentrations for Bloomsbury reveal that particle mass concentrations at this urban background site are strongly influenced by traffic emissions. This is in agreement with the substantial contribution of traffic to particulate matter in London [18]. Median enhancements of 7 µg m^3 for the $PM_{2.5}$ (representing about 33% of $PM_{2.5}$ concentrations) and 4.5 µg m^{-3} for the PM_{coarse} (representing about 45% of PM_{coarse} concentrations) are found for Marylebone Road over Bloomsbury. Very similar absolute (7.9 µg m^3) and relative (30%) enhancements are found between a street and a background sites in The Netherlands [21]. Janssen et al. [21] have attributed 80% of the increment in $PM_{2.5}$ concentrations at the street site to elemental carbon, which is in agreement with the significant contribution of diesel vehicles. The relatively higher enhancement for the PM_{coarse} fraction is consistent with the fact that this fraction of particulate matter is less easily transported in the atmosphere than the fine fraction and as a consequence, local sources will have a larger impact on PM_{coarse} concentrations.

The enhancement of particle mass at Marylebone Road due to the traffic also depends on the wind direction (Figure 3). It is at a maximum when the wind blows parallel to the street and when the southerly wind vortex is involved, that is to say when the wind direction favours the measurement of on-road emissions. A zero enhancement is found when the wind comes from northerly directions: $PM_{2.5}$ and

PM_{coarse} concentrations measured at Marylebone Road are similar to those measured in the urban background of London.

In contrast, the enhancement of particle mass at Marylebone Road is largely independent of wind speed (Figure 2). The same dependence on the wind speed is observed for both sites and for both PM fractions. Similar enhancements to those deduced from the weekly cycles are observed. These enhancements correspond to average heavy-duty and light-duty traffic of about 290 and about 3900 vehicles per hour respectively. The fact that these enhancements are largely independent of wind speed shows, in agreement with Harrison et al. [12], that the coarse particulate matter at Marylebone Road has two components, a wind-suspended component that is the same at the urban background site, and a non-wind related component corresponding to the enhancement due to Marylebone Road traffic. Similarly, the $PM_{2.5}$ has two components, one influenced by wind speed and corresponding to $PM_{2.5}$ urban background concentrations and a non-wind component corresponding to fresh fine particulate emissions less affected by the synoptic wind speed likely due to the closeness between emission sources and monitors, and the importance of the within-canyon circulation.

The enhancement of $PM_{2.5}$ concentrations is assumed to be all from exhaust emissions: $PM_{2.5}$ concentrations from abrasion processes or resuspension are assumed to be negligible in comparison to exhaust emissions. According to APEG [8], 90% of the mass of PM_{10} emitted from catalyst petrol and diesel exhaust, and 80% of the mass of PM_{10} emitted from non-catalyst petrol exhaust are in the below 2.5 µm size range. Using the national composition of UK vehicle fleet km for urban roads, an average of 87% of the mass of PM_{10} emitted from vehicle exhaust is estimated in the < 2.5 µm size range for Marylebone Road. This allows an estimation of PM_{coarse} concentrations due to non-exhaust vehicle emissions and then, to the estimation of PM_{10} concentrations from non-exhaust vehicle emissions. Since $PM_{2.5}$ in the roadside increment is considered to be all from exhaust emissions, these figures are minimum estimations.

Respectively 21.4% and 78.6% of the increment in PM_{coarse} are estimated to arise from exhaust and non-exhaust vehicle emissions (median 1.00 µg m^{-3} and 3.50 µg m^{-3} respectively). In average 31.4% (and up to 54%) of PM_{10} vehicle emissions are estimated to be non-exhaust emissions (possibly dominated by resuspension). This means that non-exhaust vehicle emissions contribute substantially to local PM_{10} concentrations.

Normalisation of the enhancements by the number of heavy duty vehicles gives contributions that are not constant (Figure 8). The same daily cycle with the lowest values during the day and the highest values during the night can be observed for both fractions of PM. These cycles are anticorrelated with the daily temperature cycle and are in agreement with the day to night evolution of the characteristics of the boundary layer: during the day, the dilution is stronger due to a higher boundary layer height and a stronger turbulence. During the stable nocturnal period, relative enhancements are up to 6 times higher than during the day for both fractions.

Other Local Influential Sources : Construction and Demolition Activities

Demolition activities from July to November 1999 and building works from February to April 2000 in the vicinity of the monitoring station influenced particle concentrations measured at Marylebone Road. Very high concentrations (above 100 µg m^{-3}) of PM_{coarse} as well as $PM_{2.5}$ have been measured over short periods. Both low and stronger wind speeds were associated with events of high concentrations and very high PM_{coarse} concentrations occurred irrespective of rainy or dry weather. Ratios of $PM_{2.5}/PM_{10}$ were generally lower than 0.5 indicating that this source had higher PM_{coarse} emissions than $PM_{2.5}$ emissions. The magnitude of the concentrations measured varied highly during these periods.

Which Parameters are Responsible for High Concentrations at Marylebone Road?

Hourly PM_{10} concentrations above 66 µg m^{-3} were examined (N = 908). They correspond to the highest 5% of hourly concentrations and are spread out over 219 days. These can contribute significantly to daily PM_{10} concentrations; 33.8% of the hourly PM_{10} concentrations above 66 µg m^{-3} occurred on days with daily PM_{10} concentrations above 50 µg m^{-3}. Conversely, 68.5% of days with daily PM_{10} concentrations above 50 µg m^{-3} have hourly concentrations above 66 µg m^{-3}. High hourly $PM_{2.5}$ concentrations (above 44 µg m^{-3}) and high hourly PM_{coarse} concentrations (above 28 µg m^{-3}) were also examined.

Hourly concentrations above 66 μg m^3 that occurred during known periods of construction or demolition activities in the vicinity of Marylebone Road supersite are examined separately. Numerous high concentrations are measured during these periods: 43% of PM_{10} concentrations above 66 μg m^{-3} and 84% of PM_{10} concentrations above 100 μg m^3. In agreement with the influence of construction or demolition activities, most of concentrations above 66 μg m^3 (80%) occurred during the working time of the workdays. Most are episodes of both high $PM_{2.5}$ and PM_{coarse} concentrations; even though the coarse fraction of particulate matter contributes highly to the loading (median ratio $PM_{2.5}/PM_{10} = 0.49$; while the median ratio for all events is 0.67). For 95.1%, high concentrations were favoured by a southerly wind vortex situation or winds blowing parallel to the street. Warm temperatures (median temperature = 16.5°C) and low relative humidity (median relative humidity = 63%) might have favoured the dispersion of fugitive dusts during these periods. Conversely, whilst rain events possibly reduce the impact of fugitive dusts, rain events do not seem to cancel the occurrence of such events. Some episodes of high PM_{10} concentrations in March 2000 were not due to building works but corresponded to the exceptional long range transport of dusts from the Sahara region of North Africa that influenced PM_{10} concentration levels in the UK [25].

Factors influencing PM_{10} concentrations above 66 μg m^3 that did not occur during periods with known demolition/construction activities have been investigated. Whilst the possible influence of roadworks cannot be discounted, it has not been investigated due to a lack of data. Some 72.2% of these events occurred when the heavy-duty traffic was heavy (midday + morning) and 50.1% during the midday period of the workday when the heavy-duty traffic is at a maximum. For 96.1% of these events, high concentrations were favoured by a southerly wind vortex situation or winds blowing parallel to the street. The events were equally distributed between these two situations. Polluted events with $PM_{10} > 66$ μg m^3 have been split between events with ratios $PM_{2.5}/PM_{10}$ below 0.5 ($R < 0.5$, the coarse fraction of particulate matter dominates) and events with ratios $PM_{2.5}/PM_{10}$ above or equal to 0.5 ($R \geq 0.5$, the fine fraction of particulate matter dominates). The same proportion of heavy-duty vehicles is found for these two kinds of events and both are associated with high concentrations of NO_x, suggesting that there are other influencing parameters. Different meteorological conditions are associated with these two kinds of events. The events with $R < 0.50$ correspond to dryer periods (median relative humidity = 58%) than events with $R \geq 0.50$ (median relative humidity = 71%, which is also the median relative humidity for all events). That suggests a possible role of the carriageway condition in dust resuspension. The events with $R \geq 0.50$ correspond to periods with weaker winds (median wind speed = 2.6 m s^{-1}) than events with $R < 0.50$ (median wind speed = 4.6 m s^{-1}).

Events of high $PM_{2.5}$ concentrations and events of high PM_{coarse} concentrations were also examined. Both have the same pattern as the events of high PM_{10} concentrations. However, the former have a much larger proportion of high hourly $PM_{2.5}$ concentrations during the night or the weekends (about 20%). Half of these high $PM_{2.5}$ concentrations are probably related to the traffic (large volume of light duty traffic) even though the heavy-duty traffic is low. They mostly occurred during the night (94%) and simultaneous very high concentrations of NO_x were measured (median 812 µg m^{-3}, which is above the percentile 0.75 for the overall dataset). This may be explained by a larger number of diesel light duty vehicles during the night (e.g. taxis). All conditions favouring the measurement of high concentrations are met (southerly vortex or wind blowing parallel to the street, low wind speed, high pressure, stable nocturnal layer). The other events of high $PM_{2.5}$ concentrations are episodes of long range transport from continental Europe, often associated with low wind speed (median 2.3 m s^{-1}) and fair weather (warm, drought periods).

The present paper has investigated $PM_{2.5}$ and PM_{coarse} ($PM_{2.5-10}$) sources, and the sources and processes affecting their concentrations in a street canyon in Central London. $PM_{2.5}$ and PM_{coarse} concentrations were examined in relation to the relative flows of light and heavy-duty traffic (on average 90.5% and 9.5% respectively) in Marylebone Road. The averaged particle data measured at Marylebone Road show daily and weekly cycles. Their variations are in good agreement with the traffic counts and correlate more closely with the heavy-duty traffic counts.

Some pollutant dispersion mechanisms are found to affect the two fractions of particulate matter similarly. Particle concentrations depend strongly on the prevailing wind direction which is an effect of the microscale meteorology created by the buildings surrounding Marylebone Road. A vortex situation inside the street canyon leads to high concentrations for perpendicular southerly winds (measurements strongly influenced by on-road emissions) and concentrations similar to those of the urban background of

London for winds in the opposite direction. High concentrations are also measured when the wind blows parallel to the street. During the stable nocturnal boundary layer the relative contributions of vehicles to particulate matter are up to six times higher than during during the daytime convective mixed boundary layer.

This study gives some insights into the sources and processes influencing $PM_{2.5}$ and PM_{coarse} concentrations in a street environment.

- **For $PM_{2.5}$** : Examination of the discriminated traffic counts at Marylebone Road has shown a strong link between the $PM_{2.5}$ and heavy-duty vehicle traffic. Heavy-duty vehicles are mainly diesel vehicles and are strong emitters of particles. The examination of emission factors reveals that lorries dominate the exhaust emissions and shape the weekly cycle of particles measured at Marylebone Road. Heavy-duty vehicles correspond on average to only 8.6% of the total traffic but contribute significantly to the atmospheric burden of fine particulate matter. It is estimated that heavy-duty vehicles are responsible for on average 62% of PM_{10} from exhaust emissions. This substantial impact of the less numerous heavy-duty traffic on airborne particulate matter was already suggested by the study of [22]. A median enhancement of $PM_{2.5}$ concentrations of about 7 µg m^{-3} due to the local traffic in Marylebone Road is evaluated for a hourly average of about 290 heavy-duty vehicles and about 3900 light duty vehicles per hour. This enhancement corresponds to one third of median $PM_{2.5}$ concentrations measured at Marylebone Road and is largely independent of the wind speed. Because $PM_{2.5}$ concentrations from the urban background are strongly influenced by the wind speed, the overall $PM_{2.5}$ concentrations decrease when the wind speed increases, reflecting the effect of the dilution and dispersion of pollutants by stronger wind speeds. Local construction and demolition activities as well as long range transport occasionally lead to high $PM_{2.5}$ concentrations at Marylebone Road.
- **For PM_{coarse}** : In contrast to the fine fraction, coarse particulate matter concentrations increase when the wind speed increases. On the other hand, a strong correlation is found between PM_{coarse} concentrations and heavy-duty traffic. This means that the coarse fraction of particulate matter at Marylebone Road is derived partly from wind-blown resuspended dusts and partly from particulate

matter resuspended or emitted by the heavy-duty traffic. The latter particulate matter is independent of the wind speed and is either resuspended road-dusts or particles from abrasion processes (tyre wear, brake linings); while, the former corresponds to the local urban background and might be PM_{coarse} from activities such as construction works or possibly from natural sources. PM_{coarse} in the urban background is also produced mechanically by the traffic, as suggested by the weekly cycle, and might be kept airborne by stronger winds. Particles induced by local Marylebone Road traffic correspond on average to 45% of PM_{coarse} measured at Marylebone Road (median enhancement of about 4 µg m^{-3} for about 290 heavy-duty vehicles per hour and about 3900 light duty vehicles per hour). It is estimated that at Marylebone Road, at least 78.6% of PM_{coarse} and at least 31.4% of PM_{10} are from non-exhaust emissions. Construction and demolition activities are responsible for occasional much larger PM_{coarse} concentrations than traffic. Coarse particles are often considered to be of natural origin (sea salt, pollen, fungal spores…). In this paper, we show that traffic as well as building work activities contribute substantially to the loading of coarse particles in an urban atmosphere.

The patterns of events surrounding high hourly particulate matter concentrations at Marylebone Road is examined. Almost half of polluted events are episodes during construction and demolition activities in the vicinity of the Marylebone Road air sampling station. The other polluted events often occur when heavy-duty vehicle traffic is high and when the prevailing wind direction favours the measurement of on-road emissions. Two kinds of events have been defined: (1) Events with high PM_{coarse} and/or ratio $PM_{2.5}/PM_{10} < 0.5$ which generally occur when the wind speed is stronger and the relative humidity lower. (2) Events with high $PM_{2.5}$ concentrations and/or ratio $PM_{2.5}/PM_{10} \geq 0.5$ that often occur during situations of poor dispersion of pollutants (low wind speed) or occasional episodes of long range transport.

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	1998	**1999**	**2000**
Motorcycles	0,088	0,088	0,088
Light vehicles	0.030	0.029	0.025
Rigid lorries	0.403	0.350	0.263
Articulated lorries	0.566	0.532	0.457
Buses/Coaches	0.775	0.573	0.451

Table 1 : Average emission factors calculated for Marylebone Road vehicle classes.

a

$\mu g\ m^{-3}$	$PM_{2.5}$	PM_{coarse}	PM_{10}
Minimum	**1**	**1**	**3**
Maximum	**330**	**595**	**800**
Median	**21**	**10**	**32**
Arithmetic mean	**23.3**	**12.7**	**36.0**

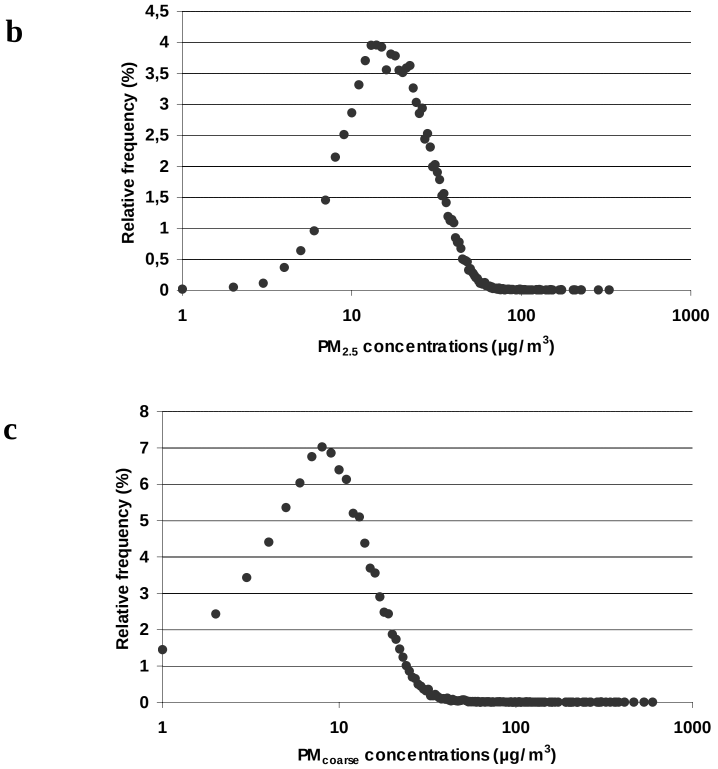


Figure 1

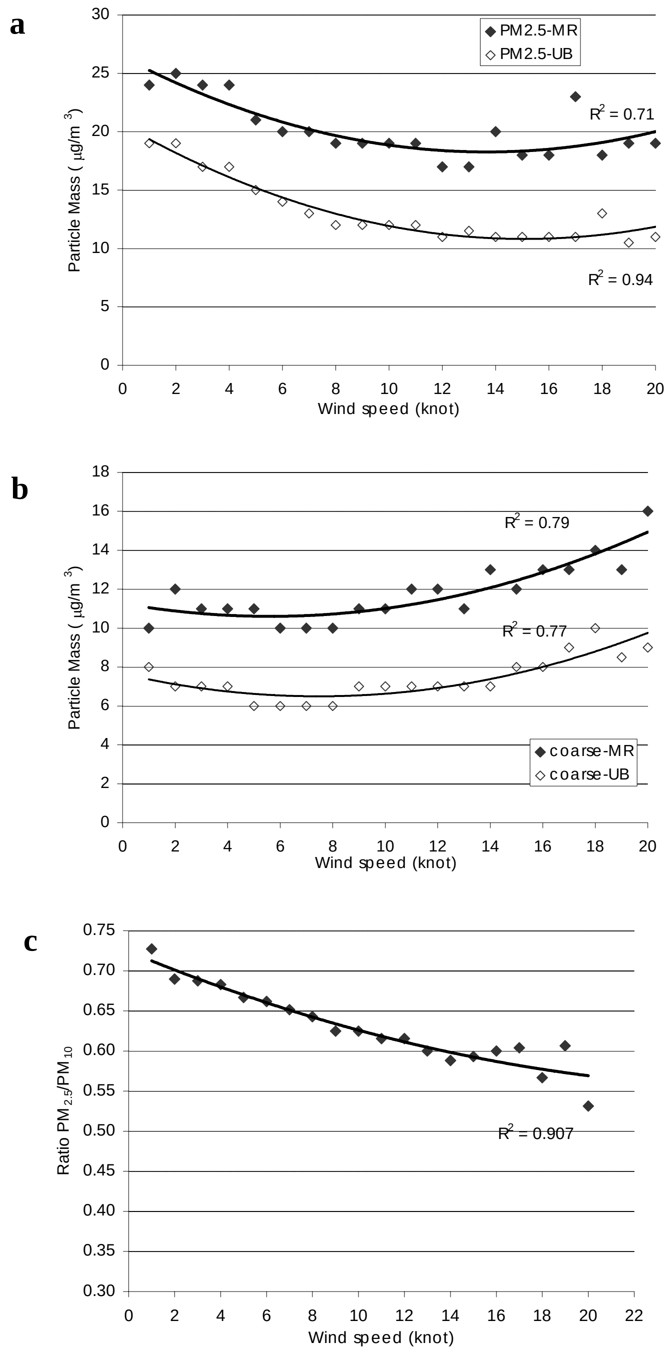

a
30
25
20
15
10
5
0
Particle Mass (μg/m 3)
◆ PM2.5-MR
◇ PM2.5-UB
R2 = 0.71
R2 = 0.94
0
2
4
6
8
10
12
14
16
18
20
Wind speed (knot)
b
18
16
14
12
10
8
6
4
2
0
Particle Mass (μg/m 3)
R2 = 0.79
R2 = 0.77
◆ coarse-MR
◇ coarse-UB
0
2
4
6
8
10
12
14
16
18
20
Wind speed (knot)
c
0.75
0.70
0.65
0.60
0.55
0.50
0.45
0.40
0.35
0.30
Ratio PM2.5/PM10
R2 = 0.907
0
2
4
6
8
10
12
14
16
18
20
22
Wind speed (knot)

Figure 2

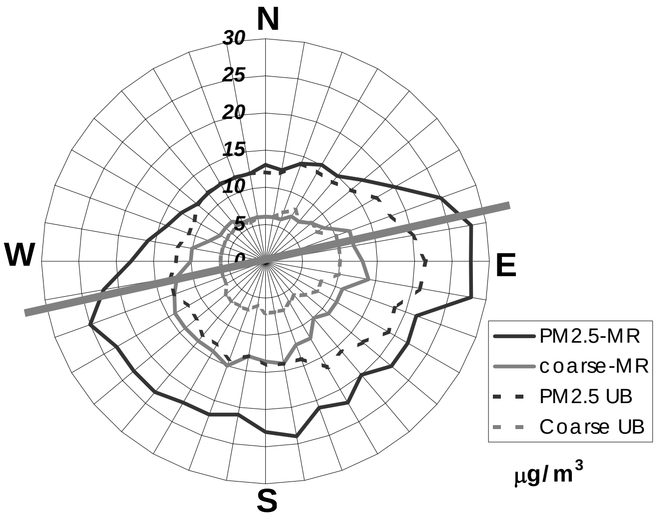


Figure 3

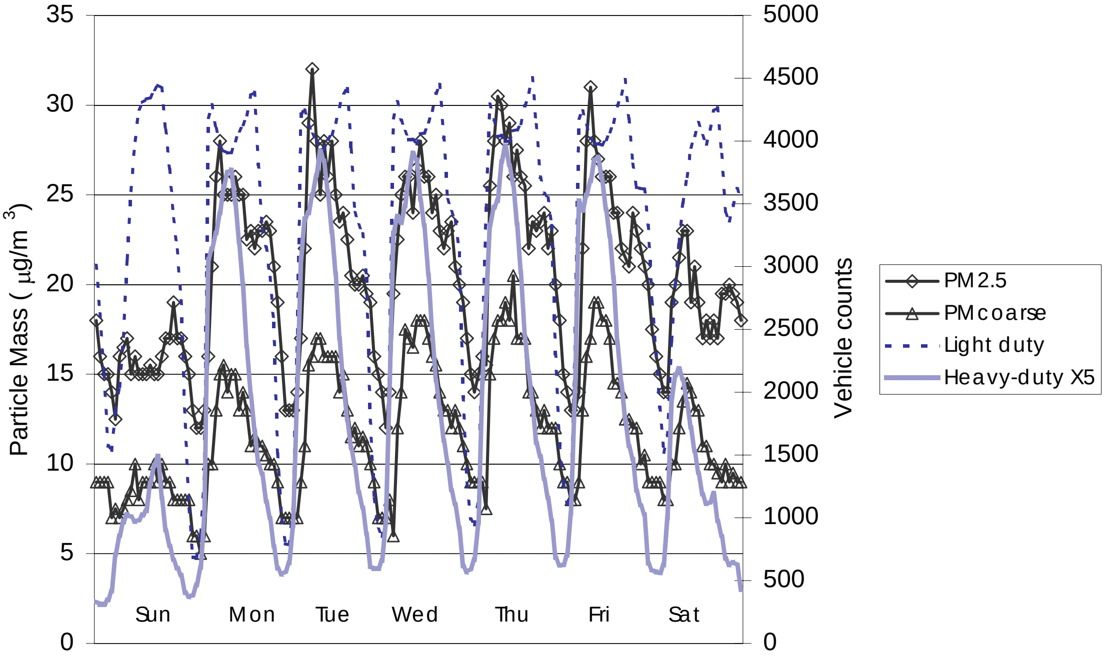

35
30
25
20
15
10
5
0
Particle Mass (µg/m 3)
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
Vehicle counts
Sun
Mon
Tue
Wed
Thu
Fri
Sat
PM2.5
PMcoarse
Light duty
Heavy-duty X5

Figure 4

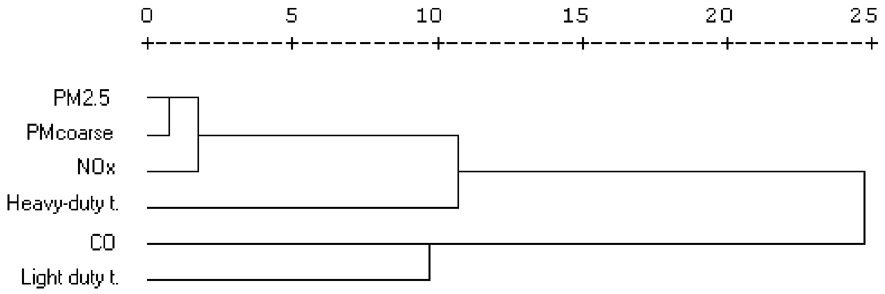
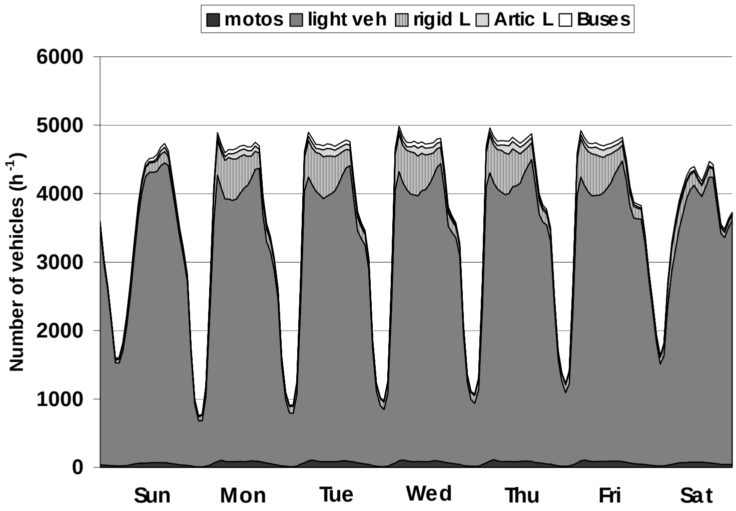


Figure 5

a


motos
light veh
rigid L
Artic L
Buses
Number of vehicles (h^{-1})
6000
5000
4000
3000
2000
1000
0
Sun
Mon
Tue
Wed
Thu
Fri
Sat

b

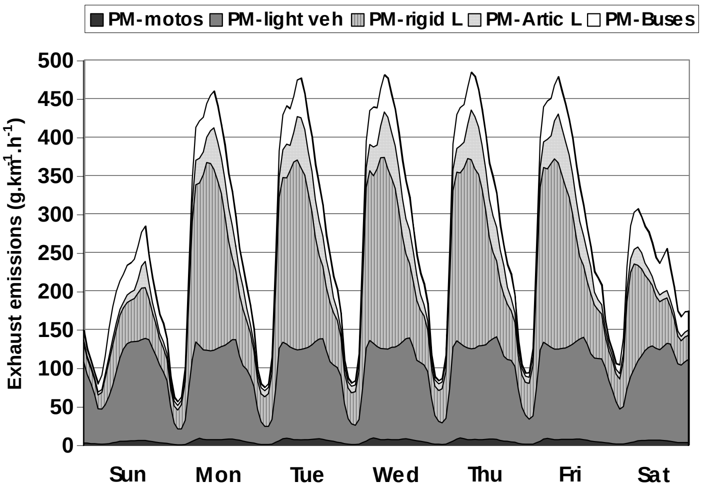

PM-motos
PM-light veh
PM-rigid L
PM-Artic L
PM-Buses
Exhaust emissions ($g.km^{-1}.h^{-1}$)
500
450
400
350
300
250
200
150
100
50
0
Sun
Mon
Tue
Wed
Thu
Fri
Sat

Figure 6

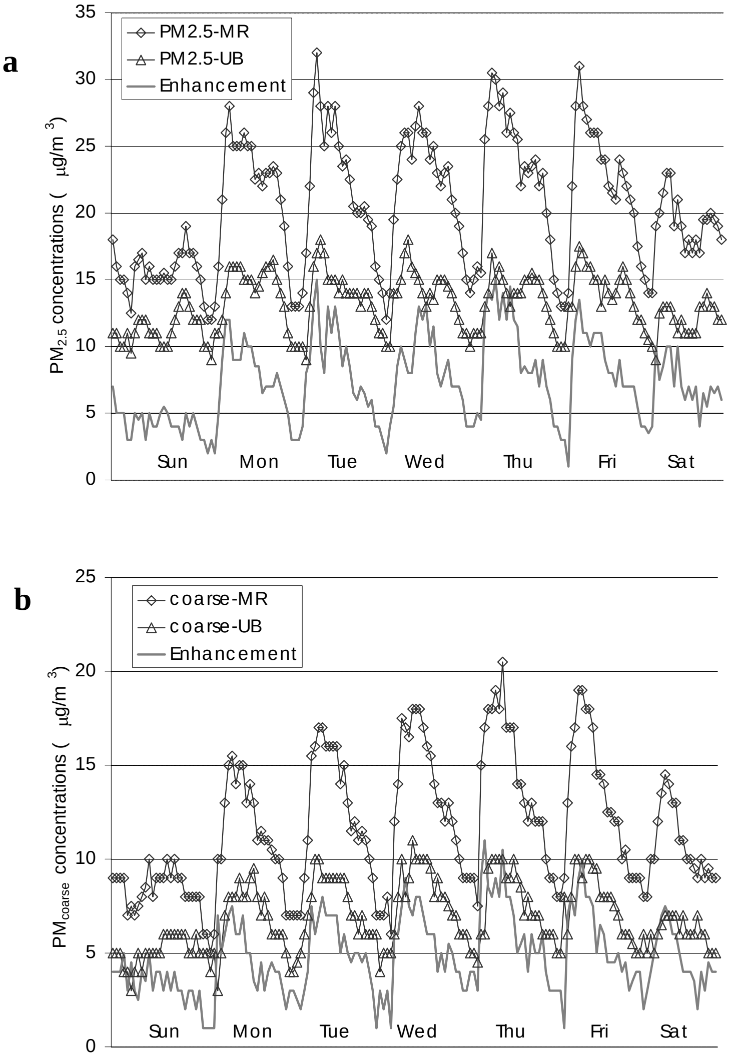
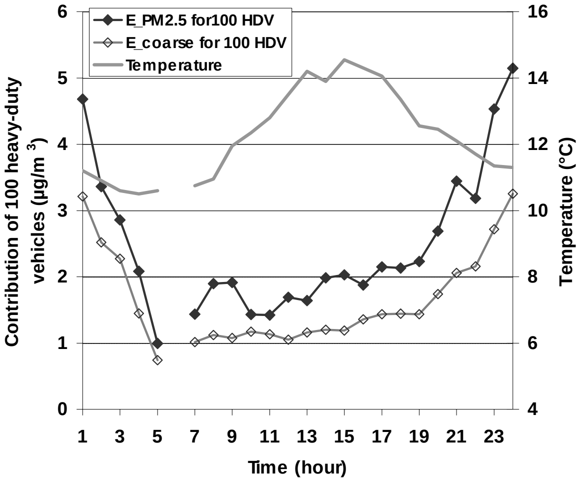

a
PM2.5-MR
PM2.5-UB
Enhancement
PM$_{2.5}$ concentrations (µg/m^3)
35
30
25
20
15
10
5
0
Sun
Mon
Tue
Wed
Thu
Fri
Sat
b
coarse-MR
coarse-UB
Enhancement
PM$_{coarse}$ concentrations (µg/m^3)
25
20
15
10
5
0
Sun
Mon
Tue
Wed
Thu
Fri
Sat

Figure 7



E_PM2.5 for100 HDV
E_coarse for 100 HDV
Temperature
Contribution of 100 heavy-duty vehicles (µg/m 3)
Temperature (°C)
Time (hour)
0
1
2
3
4
5
6
4
6
8
10
12
14
16
1
3
5
7
9
11
13
15
17
19
21
23

Figure 8