Atmospheric Environment 44 (2010) 5139–5147

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Vertical transport of accumulation mode particles between two street canyons and the urban boundary layer

J.A. Salmond [a,*], L. Pauscher [a,1], G. Pigeon [b], V. Masson [b], D. Legain [b]

[a] School of Environment, University of Auckland, Private Bag 92019, Auckland Mail Centre, Auckland 1142, New Zealand
[b] Centre National de Recherches Meteorologiques, Meteo-France, Toulouse, France

ARTICLE INFO

Article history:
Received 3 March 2010
Received in revised form
17 August 2010
Accepted 1 September 2010

Keywords:
Turbulent fluxes
Accumulation mode particles
Urban canopy layer
Urban boundary layer
Particle fluxes
Toulouse France

ABSTRACT

Concentrations and turbulent fluxes of accumulation mode particles were measured during the 2004–2005 ‘Canopy and Aerosol Particle Interaction in Toulouse Urban Layer’ project (CAPITOUL) at the top of two intersecting street canyons and in the urban boundary layer (UBL) in Toulouse, France. Particle numbers were strongly affected by boundary layer depth and showed limited sensitivity to local emissions. Differences in the diurnal patterns of particle numbers were observed between the finer fraction (0.3–0.4 μm) and coarser fraction (1.6–2.0 μm) of accumulation mode particles, indicating different processes of formation, evolution and transportation may be dominant. Highest particle numbers were observed in the narrow street canyon which had more limited local emissions and comparatively small particle fluxes. However, the improved ventilation rate in the wider canyon was also associated with the downward mixing of particles into the street canyon from the UBL. The results from this study clearly illustrate the temporal and spatial variability of particle numbers and fluxes in the urban atmosphere.

1. Introduction

High concentrations of particulate matter (PM) in the atmosphere have repeatedly been shown to have significant impacts on human health (Dockery et al., 1993; Wilson et al., 2001; Laden et al., 2006; Zeger et al., 2008). In urban environments horizontal dispersion is limited by building configurations below roof tops in the urban canopy layer (UCL) (Vardoulakis et al., 2003). As a consequence the ventilation provided by vertical transport processes may play a significant role in determining street level concentrations of PM. Vertical transport processes also provide an important contribution to the net emission of PM from urban areas (Martensson et al., 2006). Measurements of the rate of vertical exchange of PM in the UCL and the urban boundary layer (UBL) aloft therefore have the potential to improve our understanding of the impact of PM on regional and global climate change processes (IPCC, 2001; Liu and Daum, 2002; Martensson et al., 2006; Calvo et al., 2008; Gomes et al., 2008).

A range of different size particles are observed in the urban atmosphere. Three types of fine particles may be identified: nucleation and Aitken mode (which collectively are known as ultrafines and are <0.1 μm in diameter) and accumulation mode particles (0.1–2.5 μm) (Hussein et al., 2005; Birmili et al., 2001). One mode of coarse particles (>2.5 μm) is also identified (Bloss, 2009). This study focuses on the vertical exchange processes which govern particulate transport in urban environments. Accumulation mode particles were chosen for this study as ultrafine particles can show very rapid rates of particle formation which approach those of turbulent transport, thereby confounding flux measurements. Similarly, the smaller number of particles in the coarse size fraction limits the statistical rigor of the analysis of particle fluxes.

There are many sources of accumulation mode PM at different heights within the urban atmosphere. For example, they may be resuspended or emitted locally at the surface from diesel exhaust emissions (Morawska et al., 1998) and abrasion processes (Aatmeeyata and Sharma, 2009), emitted directly into the UBL from domestic heating, industrial and commercial chimney sources (such as kitchens), transported into the urban region by advection processes, or formed in the urban atmosphere as a result of physico-chemical changes such as coagulation and condensational growth (Charron and Harrison, 2003).

Unlike nucleation mode particles or coarse particles, accumulation mode particles have a longer residence time in the atmosphere, determined primarily by the frequency of precipitation events (Bloss, 2009). Once in the atmosphere particles are subject

---

* Corresponding author. Tel.: +64 9 3737599x88650; fax: +64 9 3737434.
E-mail address: J.salmond@auckland.ac.nz (J.A. Salmond).

[1] Present address: Environmental Monitoring and Modelling Group, Department of Geography, King’s College London, The Strand, London, WC2R 2LS, UK

1352-2310/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2010.09.002

to mean and turbulent transport processes (Bauman et al., 1982; Vakeva et al., 1999; Wehner and Wiedensohler, 2003). The complexity of the underlying local and regional emissions, transport and formation processes result in strong inter and intra site variability (Wilson et al., 2005, Pakkanen et al., 2003; Weber et al., 2006). For example, some previous studies have shown weak correlations of accumulation mode particles with traffic and meteorological parameters (Pakkanen et al., 2003; Weber et al., 2006) while others report stronger correlations (e.g. Hussein et al., 2006). Variations in concentration with height are also reported with some studies identifying increased concentrations near the surface explained by the dominance of local sources (Morawska et al., 1999) and other reporting higher concentration aloft or no variation with height, both explained by high rates of long range transport and subsequent mixing downwards into the street canyon (Colls and Micallef, 1999; Wehner and Wiedensohler, 2003; Park et al., 2004).

Some of the variability reported may be explained by the different characteristics of particle size fractions within the accumulation mode category. For example Pakkanen et al. (2003) argue that in Helsinki concentrations of the 0.15–0.4 μm size fraction is determined by local processes, whilst 0.4–1.3 μm size fraction predominantly comes from long range transport processes. Longley et al. (2004) also show a difference in the behaviour of particles in these two accumulation mode size bands. They conclude that whilst the smaller 0.1–0.5 μm size band observed in Manchester show minimum concentrations pre-dawn and a marked peak coinciding with the morning traffic flows, the larger 0.5–2.0 μm band shows a more complex diurnal distribution.

An improved understanding of turbulent fluxes of PM is vital to determining the dispersion of particles in an urban area. However, very few studies have examined turbulent fluxes of particles (in any size range) either within the UCL or the UBL above. Longley et al. (2004) provide some of the only measurements of accumulation mode particle fluxes within the UCL. The results showed a marked diurnal cycle peaking during the day. This study however was limited to two weeks in October, measurements were only made at one site (though detailed variation with height are reported) and did not include simultaneous measurements in the UBL.

This paper presents the results of a study carried out during the Canopy and Aerosol Particle Interaction in Toulouse Urban Layer (CAPITOUL) project in 2004–5 (Masson et al., 2008). Particle fluxes were measured using the eddy covariance approach in the UBL and at the top of two intersecting street canyons with very different geometries, orientations and emissions characteristics. The aim of the study is to document for the first time the spatial variability of accumulation mode particles near roof top level within the UCL at diurnal and seasonal scales. This study also compares fluxes of accumulation mode particles within the UCL with those observed in the UBL, providing the first simultaneous measurements of particle fluxes within the two layers. Finally the temporal and spatial variability of the emission rate of accumulation mode particles from the UCL is calculated to ascertain whether the surface is acting as a net source or sink of particles. Due to limited resources, this field study was not designed to provide a systematic survey of the differences in fluxes associated with different street geometry and orientation, but rather to examine two contrasting street canyons to evaluate the magnitude and direction of any differences observed within the UCL.

2. Methodology

2.1. Site description

Toulouse is an inland town located in the south-western part of France (Fig. 1) (Pigeon et al., 2007). It is predominantly characterised by deep, narrow street canyons irregularly orientated and interspersed with several wider canyons which act as the main

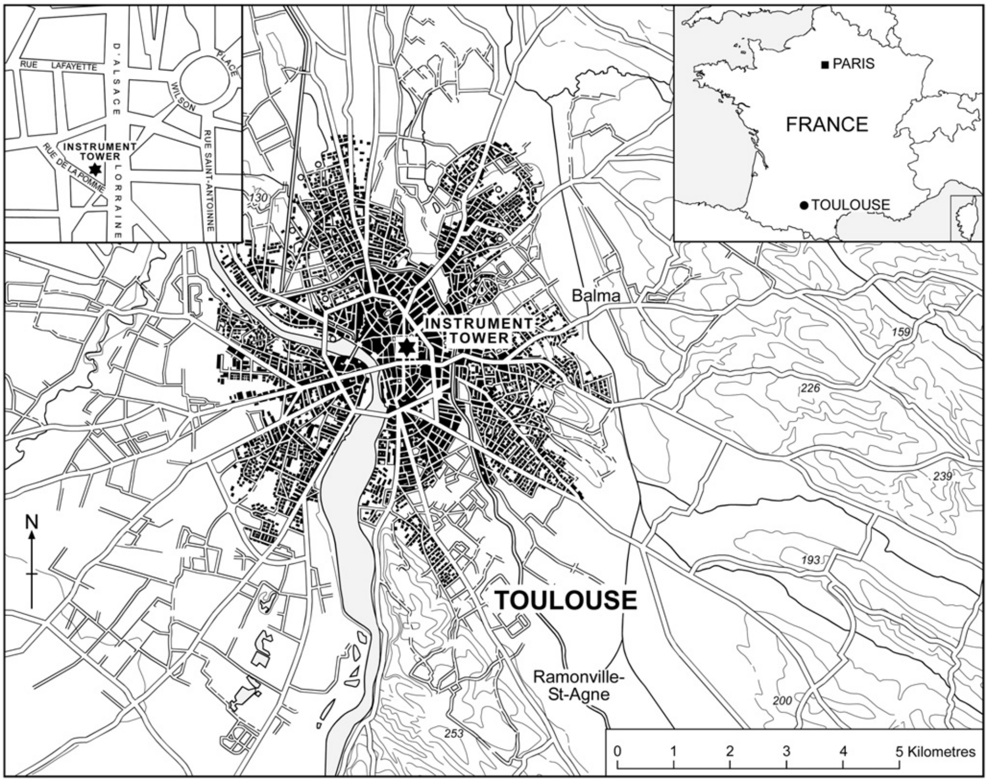

Fig. 1. Map of Toulouse. Inset shows position of tower, Rue d'Alsace-Lorraine and Rue de la Pomme.

arteries for traffic. Prevailing wind flows in Toulouse are either NW or SE (Masson et al., 2008). Mean annual temperatures vary between 4.8 °C in winter and 22.4 °C in summer (Pigeon et al., 2007). Mean annual precipitation is 656 mm, with more precipitation falling in summer than in winter (Pigeon et al., 2007).

The specific intersection studied in Toulouse is Rue de la Pomme and Rue d'Alsace-Lorraine. Rue d'Alsace-Lorraine, oriented North-South, is a main artery (and bus route) out of the city and is three lanes wide (Fig. 1). It has an aspect ratio of approximately 0.85. Rue de la Pomme is a narrower street oriented Northwest-Southeast (Fig. 1). It has only a single lane of traffic (with an additional lane for parking). It has an aspect ratio of approximately two (Masson et al., 2008). Mean building height in this area is 20 m (Masson et al., 2008) but neither street is perfectly symmetrical in cross-section.

2.2. Instrumentation

Turbulence data

The following sections describe instrumentation and data collection relevant to the present study. For a detailed description of all CAPITOUL methods refer to Masson et al. (2008). In the UBL instruments were mounted on a 27.5 m pneumonic mast (Hilomast NX30) that was extended vertically from a 20 m high roof top on the northwest corner of the intersection (Masson et al., 2008). Data analysed in this study were restricted to measurements taken when the mast was in its highest position. During the observation periods the stability was assessed using Monin–Obukhov similarity theory (z'/L where L is the Obukhov length) which ranged from neutral to unstable. Under these conditions the calculated fetch for the tower was consistently within 500 m of the measurement site (Pigeon et al., 2007). Within this region the surface characteristics of the city are relatively homogenous.

In the UCL, booms were mounted at the top of each street canyon extending one third of the way across each canyon. A 5 m boom was attached to the West wall (E-facing) over Rue d'Alsace-Lorraine and a 3 m boom was attached to the NE wall (SW-facing) over Rue de la Pomme. Given that booms extended from only one wall, and were closer to the wall on which they were mounted, they may not be representative of flows from or within the canyon under all conditions. Stability in the Rue de la Pomme ranged from neutral to unstable and in Rue d'Alsace-Lorraine from weakly stable to unstable. Stable conditions were infrequently observed during at night when winds were from the SE. The fetch for these sensors is assumed to be contained within each street canyon.

The tower and booms were instrumented with turbulence sensors and optical particle counters (GRIMM 1.108). Particle concentrations were measured at a frequency of 1 Hz. The air was drawn through a 1 m polished copper tube before reaching the particle counter resulting in a time lag of 1 s. Three dimensional wind speed and temperature were measured using a 50 Hz sonic anemometer (Gill HS 50) in the UBL and two 10 Hz sonic anemometers (RM Young) on the booms. Water vapour and carbon dioxide concentrations were measured using infrared gas analysers (Licor 7500). All the instruments were sampled and recorded by two data acquisition computers. The instruments were inter-calibrated at the beginning and end of field work, and showed no offset or drift.

Turbulence data were collected from March 2004 to February 2005. However, PM was only sampled during intensive observation periods (IOPs) during the summer (July and August, 2004) and winter months (January and February, 2005). For the purposes of this data these two IOPs have been analysed in order to make seasonal comparisons. During this period there was some data loss due to precipitation.

Few studies have discussed the need for standard corrections for the eddy covariance method for particle fluxes (Buzorius et al., 1998; Pryor et al., 2008). In this study special consideration was given to the PM data as a consequence of the non-ideal instrumental set-up. Spikes were removed from the turbulence data by applying a band of 3.5σ (Højstrup 1993), a linear regression used to remove long term trends (Gash and Culf, 1996) and standard Webb correction and stationarity tests were applied to all fluxes including particulate fluxes (Foken and Wichura, 1996). Following the theoretical considerations proposed in Leuning and Judd (1996) and Aubinet et al. (2000) the impact of sensor separation on the resulting fluxes was calculated to be negligible. Analysis of CO_2 fluxes at 10 Hz and at 1 Hz showed that errors associated with the slow sampling rate of particles fluxes were very small. Emission velocities were calculated following Longley et al. (2004).

Boundary layer depth

During the IOPs radiosondes (Vaisala RS92) were launched from the city centre about 500 m from the tower measurement site approximately 5 times per 24 hour period with a vertical resolution of approximately 5 m. Due to local time changes, releases occurred at different times during the summer and winter. BL depth was assessed using four different indicators:

a) the maximum relative humidity level below 5000 m (Marsik et al., 1995; Seibert et al., 2005)
b) the maximum of the second derivative of the potential temperature between 200 and 5000 m (Sullivan et al., 1998)
c) the minimum of the second derivative of the water vapour mixing ratio between 200 and 5000 m (Couvreux et al., 2005)
d) the lowest point below 5000 m where the virtual potential temperature is greater than the average virtual potential temperature of the levels below (Marsik et al., 1995; Seibert et al., 2005)

The results were compared and the lowest returned value chosen as indicative of boundary layer depth. A visual check was performed to verify the estimates. Data were available from 6 days during the summer IOP and 7 days for the winter IOP.

Traffic data

Two road traffic counters were located within a 500 m radius of the measurement site on Rue d'Alsace-Lorraine and Rue Lafayette. All the available hourly data for July 2004 and January 2005 were averaged to provide mean daily traffic flows.

3. Results & discussion

3.1. Diurnal trends in mean particulate numbers

Throughout the year mean concentrations of PM in both the UCL and UBL show a marked sensitivity to boundary layer (BL) depth (Figs. 2 and 3). Peak particle numbers are observed during the night (when the BL is shallow) and a rapid decrease in numbers is observed during the morning period consistent with the growth of the BL (Fig. 3a–d). Further, during the winter when maximum BL depths are approximately 10% of the summer value particle numbers are up to six times higher (Figs. 2 and 3a–d) (though increased emissions from space heating and chimneys are also expected at this time (Pigeon et al., 2007; Calvo et al., 2008)). Despite this, the diurnal decrease in concentrations in both seasons is about 50% of the nocturnal maxima. If dilution effects alone accounted for the observed diurnal variation in concentrations we might expect to see much lower daytime concentrations in the summer than currently observed. This suggests that additional

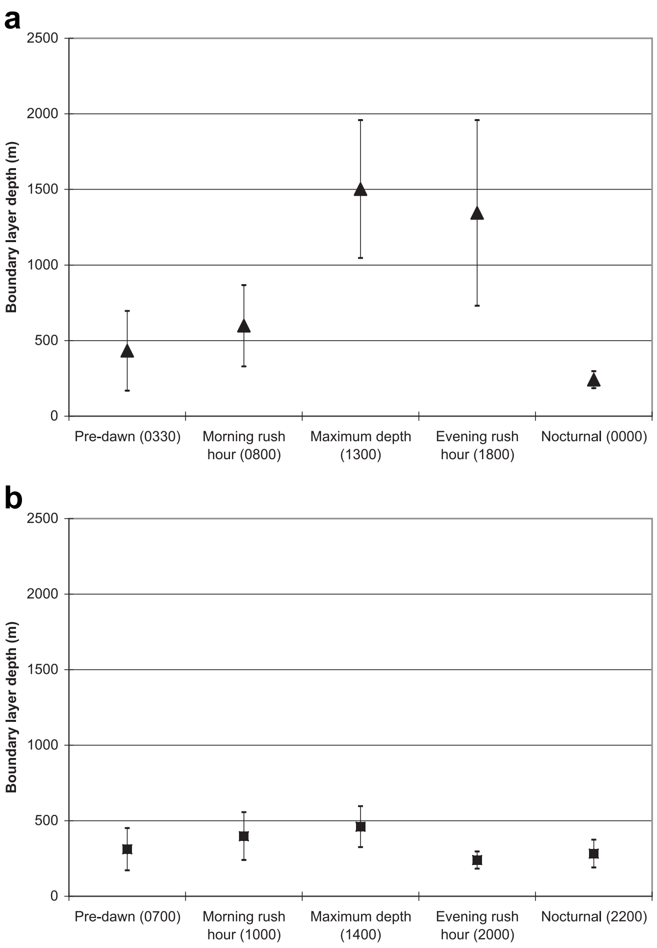

Fig. 2. Mean boundary layer depths observed during a) the summer IOP and b) the winter IOP.

particle formation, evolution and transportation processes may be active during the summer daytime period.

Gomes et al. (2008) and Calvo et al. (2008) also note the strong influence of boundary layer depth on aerosol concentrations in the Toulouse region across a wide range of particle sizes. These diurnal trends are consistent with other particle studies in regions where long range transport of particles has been identified as important and meteorological variables a strong determinant of concentrations such as those reported (e.g. Hussein et al., (2006) and Nicolás et al., (2009)). However, in Toulouse, significant correlations were not observed between particle number and wind speed, temperature or stability during either the summer or winter months. Increased numbers of accumulation mode particles were associated with NW winds (during the summer concentrations increased by 150% in the UBL, 143% in Rue d'Alsace-Lorraine and 142% in Rue de la Pomme, whilst in the winter all sites increased by 137%) when compared to winds from the SE. Given the low number of samples from other wind directions a pollution wind rose could not be identified, however the consistency of reporting between sites suggests a non-local source may account for this trend.

Despite the dominance of boundary layer effects, diurnal trends associated with local emissions were also identified in the Toulouse data set. For example, during the summer months a traffic-related increase is observed in both the finer (0.3–0.4 μm) and coarser (1.6–2.0 μm) particle size fractions between 0400 and 0600 UTC in the UCL (Figs. 3a,b and 4).

Interestingly, during the summer increased particle numbers during the morning rush hour are only reported in the UBL in the coarser fraction. Also, particle numbers of both size fractions are higher in the UCL than in the UBL during the daytime period. This suggests that the smaller size fraction may be sensitive to different particle sources, transport and/or evolution processes operating only in the UBL. During the evening period, a distinct traffic-related rush hour is not observed in Toulouse.

During the winter months a traffic-related increase in concentrations is observed between 0500 and 0700 UTC in both the UBL and UCL in both size fractions, but especially the coarser fraction where numbers are almost doubled (Fig. 3c,d and 4). During this season sunrise occurs after the main increase in traffic flows at 0730 UTC so we might expect to see a larger effect from any local surface based sources due to the shallower BL. The smaller increase in numbers of particles in the 0.3–0.4 μm finer fraction may reflect the seasonally higher numbers of particles in this size fraction from space heating emissions. Further, numbers of particles in this size fraction were consistently higher in the UBL than UCL. (Particulate data for Rue de la Pomme was unavailable for the winter period.) These trends are consistent with the increased dominance of space heating emissions. Charron and Harrison (2003) identify a decrease in concentrations of accumulation mode particles during periods of high emissions of nano-particles. This effect was not observed in Toulouse.

During both seasons, concentrations of 0.3–0.4 μm size PM in the UBL were higher than in Rue d'Alsace-Lorraine during the night (Fig. 3a,c). This suggests that during the night there is only weak coupling of the UBL with the UCL. Transport, emission, particle formation processes or reduced mixing depths affecting only the UBL allow particle numbers to build up in the air aloft, and the absence of efficient vertical transport processes inhibits the mixing of these particles down into the UCL below. (This is discussed further in later sections.)

Unexpectedly, concentrations of 0.3–0.4 μm size PM were higher in Rue de la Pomme than both those observed in the UBL and Rue d'Alsace-Lorraine throughout the summer diurnal cycle. Previous studies have shown that when prevailing wind flows are perpendicular to a street canyon (as they were along Rue d'Alsace-Lorraine during this field experiment) flow isolation can lead to much higher concentrations of fine PM (Longley et al. 2004). However, stagnant air flows have also been shown to favour increased particle numbers through reduced vertical dispersion and promotion of particle to particle coagulation and particles growth due to condensation of semi-volatile gases (Pey et al., 2008; Nicolás et al., 2009). This may be the case for Rue de la Pomme. Certainly, the diurnal cycle shows a lag between the start of the morning dilution in Rue d'Alsace-Lorraine and Rue de la Pomme and the dilution effect is less pronounced. Further, given the lighter traffic flows and the absence of any additional local sources in Rue de la Pomme it is unlikely that primary emissions account for increased particle numbers. It is interesting to note that the apparent storage of particles observed in Rue de la Pomme was less marked in the larger size class. Indeed, during the summer nocturnal periods concentrations approach those observed in the UBL and during the daytime period are very similar to those observed in Rue d'Alsace-Lorraine.

3.2. Particle fluxes

During the summer months particle number fluxes (calculated for the 0.3–0.4 μm size fraction only) show a marked diurnal cycle in the UCL and UBL (Fig. 5a). Despite lower particle numbers, higher fluxes and emission velocities were observed in Rue d'Alsace-Lorraine compared to Rue de la Pomme during the afternoon period

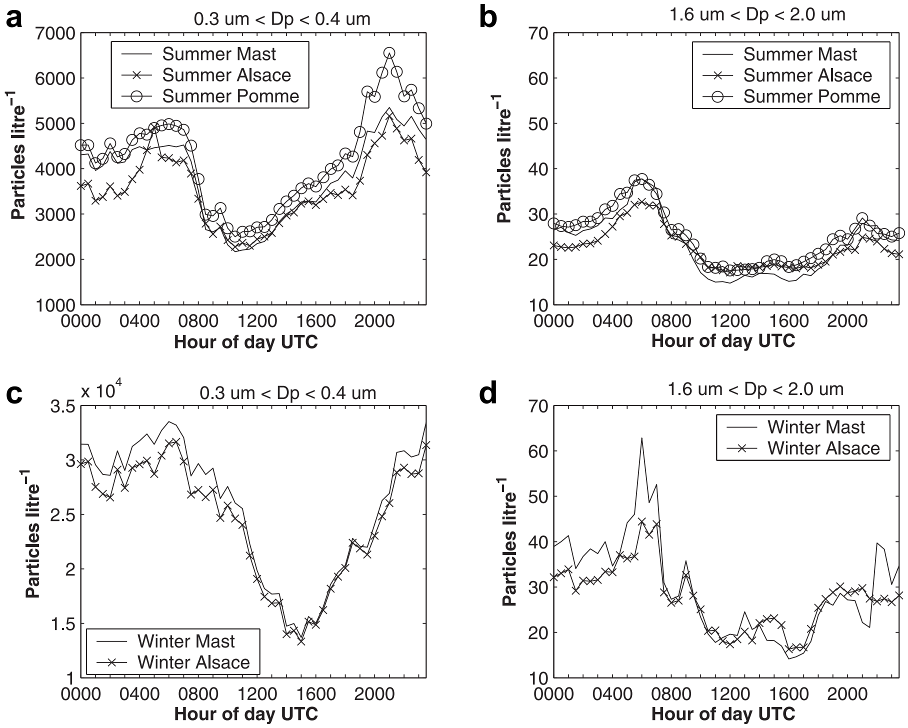

Fig. 3. Diurnal variability in particle number for a) 0.3 μm < Dp < 0.4 μm and b) 1.6 μm < Dp < 2.0 μm during the summer and for c) 0.3 μm < Dp < 0.4 μm and d) 1.6 μm < Dp < 2.0 μm size fraction during the winter IOPs.

(Fig. 6a) suggesting that vertical mixing processes are less efficient from this narrow street canyon. Further, although particle fluxes from Rue de la Pomme and Rue d'Alsace-Lorraine are very similar during the nocturnal period, on average nocturnal emissions from Rue d'Alsace-Lorraine are higher than those observed in Rue de la Pomme (Fig. 6a) and sometimes twice as large. Given the higher numbers and rapid build up particles in Rue Pomme (especially in the early evening) it is likely that particle fluxes from Rue de la Pomme are also limited by energetics during this period.

In both the UBL and UCL particle fluxes are affected by wind direction. In the UBL larger fluxes are associated with winds from the SE (Fig. 7a). Two localised peaks in the flux are observed during the summer at 1100 UTC and between 1800 and 2100 UTC (Fig. 7a). These are not related to peaks in local traffic flows and are not mirrored in the UCL fluxes suggesting that surface sources are unlikely. It is also unlikely that the peaks are accounted for by the entrainment of particles stored aloft in the growing or collapsing boundary layer, as this would be characterised by a negative (downward) flux.

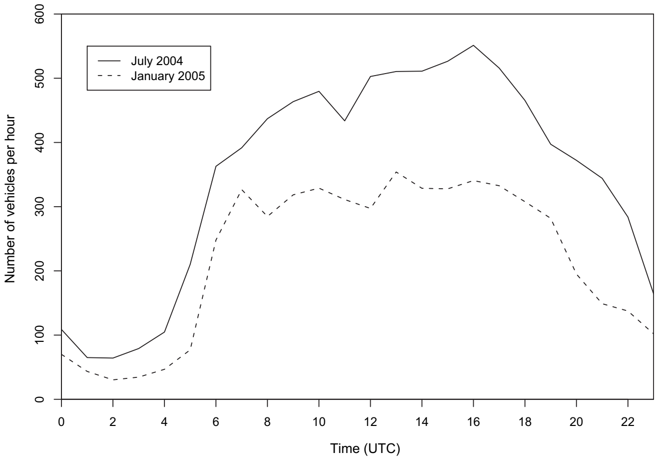

Fig. 4. Mean diurnal traffic flow volumes for two streets within 500 m of the study site in Toulouse for July 2004 and January 2005.

One factor which can affect particle growth rates is the local relative humidity. Analysis of the latent heat fluxes does show an increase in the latent heat flux in the UBL compared to the UCL in the summer during these periods (Fig. 5c) which could promote particle growth. Analysis by wind direction shows that the morning peak is independent of wind regime indicating local growth processes may be dominating (Fig. 7a). However, the evening peak is reduced and lagged by 2 h during NW winds which suggests that horizontal transport processes may become more important under this wind regime (Fig. 7a). Thus either long range transport of aged particles from elsewhere, or increased rates of situ coagulation and condensation processes localised to the UBL may account for the increased particle fluxes.

In the UCL increased fluxes are associated with NW winds. It is interesting to note that the 1100 UTC peak in particle fluxes in the UBL is coincident with the apparent drop in fluxes from Rue d'Alsace-Lorraine alone. At this time particle numbers are similar at the two heights. This switch from emission to deposition processes is a real effect in the data set which is found repeatedly on days when the winds were from the SE (Fig. 7b). This suggests that particles from aloft are being transported into Rue d'Alsace-Lorraine under these conditions. When flows are from the SE we would expect helical flows to entrain air from the UBL into Rue d'Alsace-Lorraine down the east-facing wall which is the side closest to which the instruments were mounted. When the winds are from the NW this effect is not apparent in the data set. Under these conditions flows would be expected to be exiting the street canyon up the east-facing wall loaded with particles from ground level. A more subtle

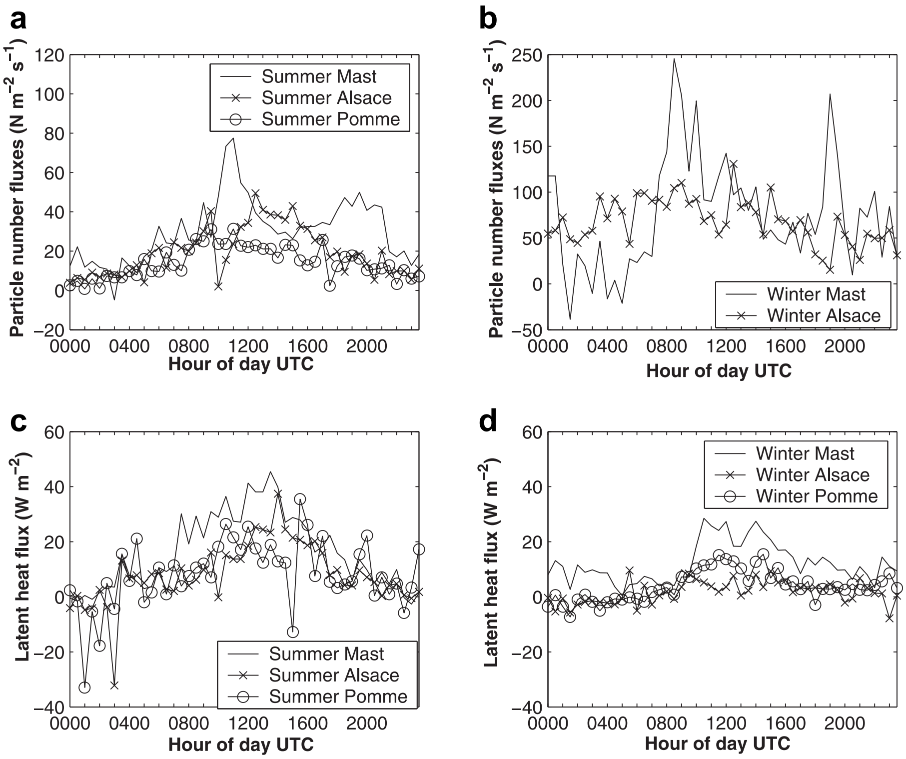

Fig. 5. Diurnal variation in 0.3 μm < Dp < 0.4 μm particle fluxes from the UBL, Rue d'Alsace-Lorraine and Rue de la Pomme during a) summer and b) winter periods and latent heat fluxes from Rue de la Pomme, Rue Alsace and the UBL during c) summer and d) winter.

reduction in fluxes from Rue d'Alsace-Lorraine can be observed in the evening period during SE wind regimes.

The absence of a coincident drop in fluxes in Rue de la Pomme (Fig. 7c) further supports the hypothesis that the coupling between the UBL and this narrower street canyon is less efficient. However, the fluxes in Rue de la Pomme do show marked differences with wind regime. Fluxes are higher during the daytime period when winds are from the SE. Although we might expect increased concentrations of particles during SE winds to be associated with the advection of particles from Rue d'Alsace-Lorraine, there is no

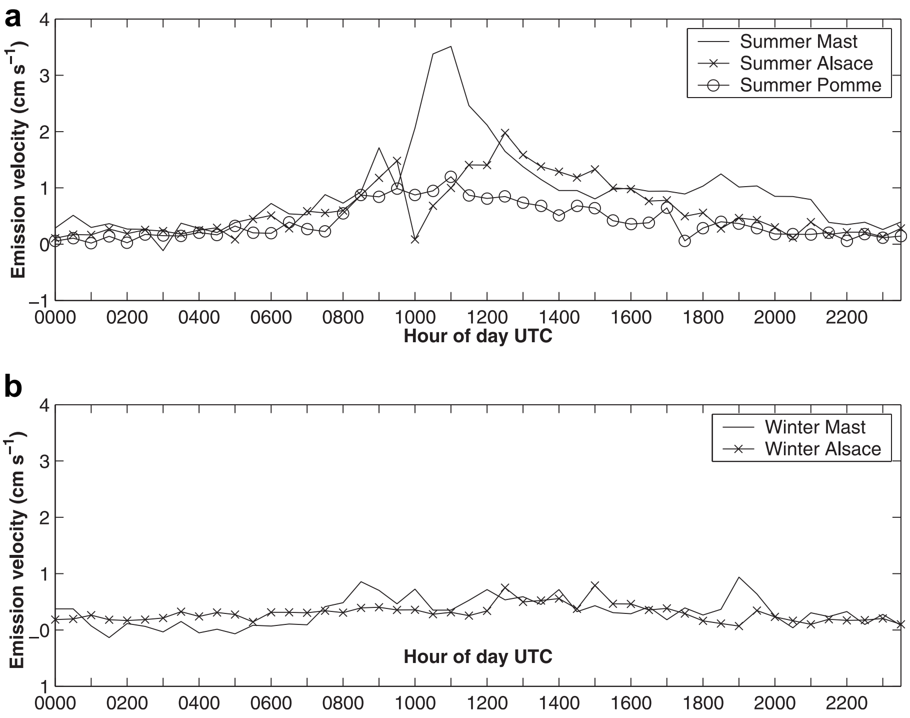

Fig. 6. Diurnal variation in emission velocities for 0.3 μm < Dp < 0.4 μm particles from the UBL, Rue d'Alsace-Lorraine and Rue de la Pomme during a) summer and b) winter.

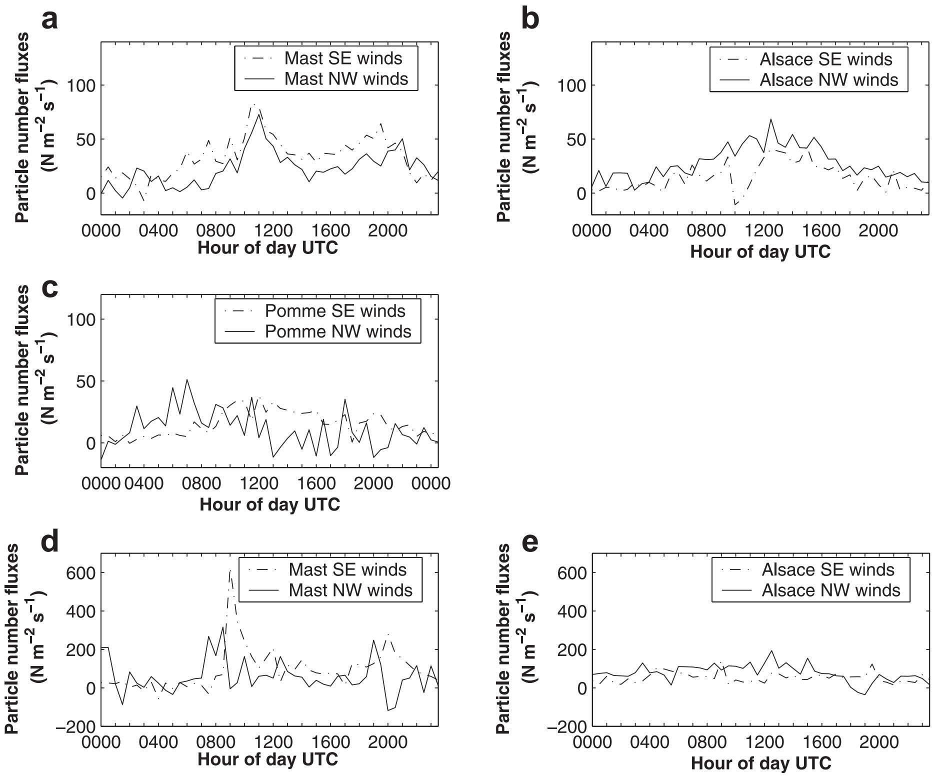

Fig. 7. Diurnal variation in 0.3 μm < Dp < 0.4 μm particle fluxes by wind regime for the summer period for a) UBL, b) Rue d'Alsace-Lorraine and c) Rue de la Pomme and winter period for d) UBL and e) Rue d'Alsace-Lorraine.

evidence for increased concentrations of PM associated with winds from this direction. Under both wind flow regimes, winds within Rue Pomme are parallel to the canyon, thus a difference in the efficiency of vertical transport was not expected. However, given the proximity of the boom to the intersection it is possible that complex flow patterns not detected with a single instrument may account for increased mixing, which in turn reduces particulate storage within the canyon accounting for reduced particle

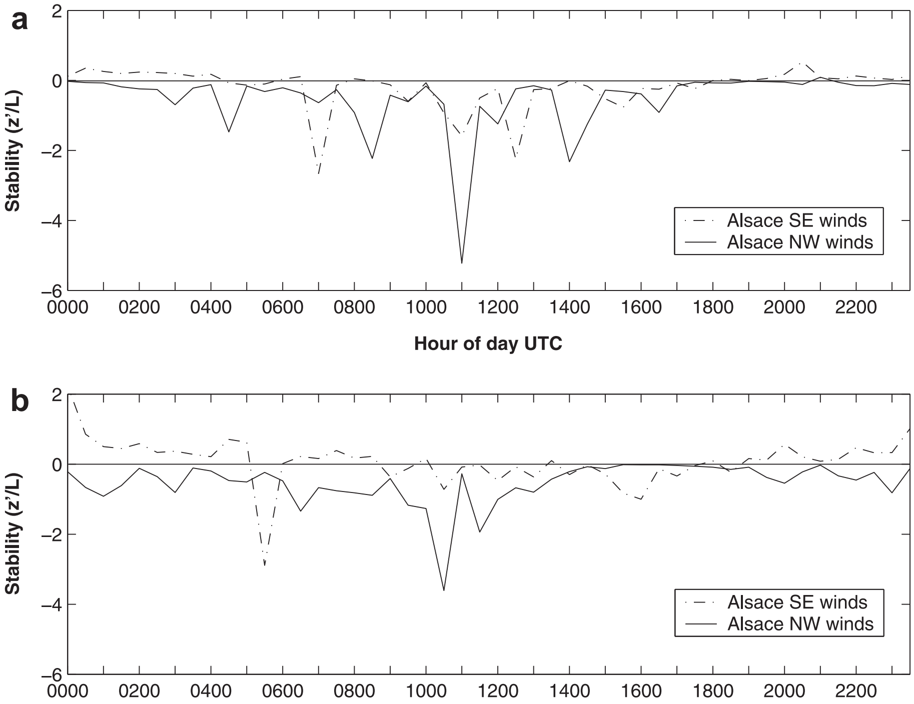

Fig. 8. Diurnal variation in stability by wind regime for Rue d'Alsace-Lorraine for a) summer and b) winter.

numbers. This needs further investigation with a detailed numerical model.

During the winter period the fluxes from both Rue d'Alsace-Lorraine and the UBL are significantly higher than during the summer months. This increase probably reflects the higher number of particles present in the atmosphere (Fig. 3) rather than improved efficiency of the vertical transport processes. In both the UBL and UCL the winter fluxes are also characterised by much greater temporal variability (Fig. 5b). Fluxes may vary by more than a factor of 4 from hour to hour and identification of clear mean diurnal trends is more challenging. This reflects the ephemeral nature of local sources such as chimney plumes, as opposed to the more homogeneous processes of accumulation and long range transport which may dominate during the summer period (Calvo et al., 2008; Gomes et al., 2008). The fluxes remain sensitive to wind direction (Fig. 7d,e).

Two distinct peaks are still discernable in the winter UBL data set, similar to the summer conditions (Fig. 5b). The first peak occurs earlier however at 0900 UTC and the second is much more temporally localised at 1900 UTC than the summer peak. A clear relationship between these peaks and latent heat fluxes is not observed. Analysis by wind direction shows a marked lag in the peaks (Fig. 7d), suggestive of horizontal transport processes rather than in-situ formation.

Interestingly a drop in the fluxes in Rue d'Alsace-Lorraine coincident with the UBL peak is not observed associated with either wind direction during the winter months (Fig. 7e). Mean number counts show that particle concentrations in the UBL and UCL do not equalise (indicative of a coupled system) until much later during the winter period (1100 UTC). Further, the Monin–Obukhov stability function for Rue d'Alsace-Lorraine indicates that unlike during the summer period, in the winter this peak in fluxes in the UBL coincides with a period when conditions in Rue d'Alsace-Lorraine are stable (Fig. 8a,b). Thus limited vertical mixing in the winter limits the transport of PM from the UBL into the UCL during the morning.

The second peak in particle fluxes in the winter in the UBL is mirrored by a small decrease in fluxes in Rue d'Alsace-Lorraine, suggesting that at 1900 UTC the sites are better coupled at this time. This is supported by particle number data (Fig. 3) and occurs despite the development of the nocturnal boundary layer.

4. Conclusions

This study has clearly illustrated the temporal and spatial variability of particle numbers and fluxes in the UCL. It has also shown the variable nature of the coupling between the two layers and the importance of vertical exchange processes in both removing particles from the surface and introducing new particles from aloft. The results demonstrate that there is a strong relation between boundary layer characteristics and accumulation mode particles in Toulouse during the summer and winter. This is consistent with other studies in urban areas where horizontal transport of particles in the UBL have been shown to be important. Thus local surface sources of primary fine PM (such as traffic-related emissions) only have a limited role in determining concentrations either in the UBL or UCL. However, like the Longley et al. (2004) study, differences in the diurnal patterns of particle numbers were observed within the size band of accumulation mode particles. This indicates that within the broad category of accumulation mode particles different processes of formation, evolution and transportation may be dominant at different size fractions.

In the UCL particle numbers were highest in the narrow street canyon (Rue de la Pomme) which had limited local emission sources and reduced particle fluxes and emission velocity were observed. This demonstrates the importance of vertical mixing processes in determining concentrations, as poor coupling to the UBL not only limits ventilation but also provides the potential for in-situ formation and accumulation of particles within the UCL.

These results have implications for pollutant exposure studies. For example, although many studies have shown reduced concentrations of gaseous pollutants such as carbon monoxide away from heavily trafficked roads (Tomlin et al., 2009), ventilation processes may reverse this effect. Further, although both emission velocities and particle number fluxes were higher in the wider, more heavily trafficked street canyon (Rue d'Alsace-Lorraine), there was also evidence for the downward mixing of particles from the UBL into the street canyon. This needs further exploration in a more detailed study which avoids the problems associated with single point measurements in the complex geometry of the UCL. More detailed modelling and experimental studies are also required to determine the local rates of accumulation mode particle production in the UBL and UCL in order to develop a more comprehensive understanding of the factors determining concentrations and emission rates from urban areas. Nevertheless the results clearly demonstrate the influence of canyon orientation and geometry in determining not only pedestrian exposure but also the impact of local surface based emissions on urban and regional climates.

Acknowledgements

This work was funded by NERC grant (NER/M/S/2003/00058) to J. Salmond and Meteo France, and was made possible due to the loan of equipment from Professor T.Oke (UBC). Thanks to Professor Ian McKendry for helpful comments on early drafts of this paper.

References

Aatmeeyata, D.S.K., Sharma, M., 2009. Traffic generated non-exhaust particulate emissions from concrete pavement: a mass and particle size study for two-wheelers and small cars. Atmospheric Environment 43, 5691–5697.

Aubinet, M., Grelle, A., Ibrom, A., Rannik, Ü., Moncrieff, J., Foken, T., Kowalski, A., Martin, S., Berbigier, P., Bernhofer, Ch., Clement, R., Elbers, J., Granier, A., Grünwald, T., Morgenstern, K., Pilegaard, K., Rebmann, C., Snijders, W., Valentini, R., Vesala, T., 2000. Estimates of annual net carbon and water exchange of forests: the EUROFLUX methodology. Advances in Ecological Research 30, 113–175.

Bauman, S.E., Williams, E.T., Finston, H.L., Ferrand, E.F., Sontowski, J., 1982. Street level versus rooftop sampling: carbon monoxide and aerosol in New York city. Atmospheric Environment 16, 2489–2496.

Birmili, W., Wiedensohler, A., Heintzenberg, J., Lehmann, K., 2001. Atmospheric particle number size distribution in central Europe: statistical relations to air masses and meteorology. Journal of Geophysical Research 106 (D23), 32005–32018.

Bloss, W., 2009. Atmospheric chemical processes of importance in cities. In: Hester, R.E., Harrison, R.M. (Eds.), Air quality in urban environments. Issues in Environmental Science and Technology, vol. 28. Royal Society of Chemistry, London, pp. 42–64.

Buzorius, G., Rannik, U., Hakela, J.M., Vesala, T., Kulmala, M., 1998. Vertical aerosol particle fluxes measured by eddy covariance technique using condensational particle counter. Journal of Aerosol Science 29, 157–171.

Calvo, A.I., Pont, V., Liousse, C., Dupre, B., Mariscal, A., Zouiten, C., Gardrat, E., Castera, P., Lacaux, C.G., Castro, A., Fraile, R., 2008. Chemical composition of urban aerosols in Toulouse, France during CAPITOUL experiment. Meteorology and Atmospheric Physics 102, 307–323.

Charron, A., Harrison, R.M., 2003. Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere. Atmospheric Environment 37, 4109–4119.

Colls, J.J., Micallef, A., 1999. Measured and modelled concentrations and vertical profiles of airborne particulate matter within the boundary layer of a street canyon. Science of the Total Environment 235, 221–233.

Couvreux, F., Guichard, F., Redelsperger, J.-L., Kiemle, C., Masson, V., Lafore, J.-P., Flamant, C., 2005. Water-vapour variability within a convective boundary-layer assessed by large-eddy simulations and IHOP_2002 observations. Quarterly Journal of the Royal Meteorological Society 131, 2665–2693.

Dockery, D.W., Ardenpope, C., Xiping, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G., Speizer, F.E., 1993. An association between air pollution and mortality in six US cities. New England Journal of Medicine 329 (24), 1753–1759.

Foken, T., Wichura, T., 1996. Tools for quality assessments of surface-based flux measurements. Agricultural and Forest Meteorology 78, 83–105.

Gash, J.H.C., Culf, A.D., 1996. Applying a linear detrend to eddy correlation data in real time. Boundary-Layer Meteorology 79, 301–306.

Gomes, L., Mallet, M., Roger, J.C., Dubuisson, P., 2008. Effects of the physical and optical properties of urban aerosols measured during the CAPITOUL summer campaign on the local direct radiative forcing. Meteorology and Atmospheric Physics 102, 289–306.

Højstrup, J., 1993. A statistical data screening procedure. Measurement Science and Technology 4, 153–157.

Hussein, T., Hämeri, K., Aalto, P.P., Paatero, P., Kulmala, M., 2005. Modal structure and spatial–temporal variations of urban and suburban aerosols in Helsinki–Finland. Atmospheric Environment 39, 1655–1668.

Hussein, T., Karppinen, A., Kukkonen, J., Harkonen, J., Aalto, P., Hameri, K., Kerminen, V.-M., Kulmala, M., 2006. Meteorological dependence of size-fractionated number concentrations of urban aerosol particles. Atmospheric Environment 40, 1427–1440.

IPCC, 2001. The Scientific Basis. Aerosols, their Direct and Indirect Effects. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change (IPCC). Climate Change 2001. Cambridge University Press, Cambridge, England.

Laden, F., Schwartz, J., Speizer, F.E., Dockery, D.W., 2006. Reduction in fine particulate air pollution and mortality extended follow-up of the Harvard six cities study. American Journal of Respiratory and Critical Care Medicine 173, 667–672.

Leuning, R., Judd, M.J., 1996. The relative merits of open- and closed-path analysers for measurement of eddy fluxes. Global Change Biology 2, 241–253.

Liu, Y., Daum, P.H., 2002. Anthropogenic aerosols: indirect warming effect from dispersion forcing. Nature 419, 580–581.

Longley, I., Gallagher, M.W., Dorsey, J.R., Flynn, M., 2004. A case-study of fine particle concentrations and fluxes measured in a busy street canyon in Manchester. UK Atmospheric Environment 38, 3595–3603.

Marsik, F.J., Fischer, K.W., McDonald, T.D., Samson, P.J., 1995. Comparison of methods for estimating mixing height used during the 1992 Atlanta Field Intensive. Journal of Applied Meteorology 34, 1802–1814.

Martensson, E.M., Nilsson, E.D., Buzorius, G., Johansson, C., 2006. Eddy covariance measurements and parameterisation of traffic related particle emissions in an urban environment. Atmospheric Chemistry and Physics 6, 769–785.

Masson, V., Gomes, L., Pigeon, G., Liousse, C., Pont, V., Lagouarde, J.-P., Voogt, J., Salmond, J.A., Oke, T.R., Hidalgo, J., Legain, D., Garrouste, O., Lac, C., Connan, O., Briottet, X., Lacherade, S., Tulet, P., 2008. The canopy and aerosol particles interactions in Toulouse urban layer (CAPITOUL) experiment. Meteorology and Atmospheric Physics 102, 135–157.

Morawska, L., Thomas, S., Bofinger, N., Wainwright, D., Neale, D., 1998. Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size distribution and correlation with gaseous pollutants. Atmospheric Environment 32, 2467–2478.

Morawska, L., Thomas, S., Gilbert, D., Greenaway, C., Rijnders, E., 1999. A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road. Atmospheric Environment 33, 1261–1274.

Nicolás, J.F., Yubero, E., Pastor, C., Crespo, J., Carratalá, A., 2009. Influence of meteorological variability upon aerosol mass size distribution. Atmospheric Research 94, 330–337.

Pakkanen, T.A., Kerminen, V.M., Loukkola, K., Hillamo, R.E., Aarnio, P., Koskentalo, T., Maenhaut, W., 2003. Size distributions of mass and chemical components in street-level and rooftop PM1 particles in Helsinki. Atmospheric Environment 37, 1673–1690.

Park, S.K., Kim, S.D., Lee, H., 2004. Dispersion characteristics of vehicle emission in an urban street canyon. Science of the Total Environment 323, 263–271.

Pey, J., Rodríguez, S., Querol, X., Alastuey, A., Moreno, T., Putaud, J.P., Dingenen, R.V., 2008. Variations of urban aerosols in the western Mediterranean. Atmospheric Environment 42, 9052–9062.

Pigeon, G., Legain, D., Durand, P., Masson, V., 2007. Anthropogenic heat release in an old European agglomeration (Toulouse, France). International Journal of Climatology 27, 1969–1981.

Pryor, S.C., Larsen, S.E., Sørensen, L.L., Barthelmi, R.J., 2008. Particle fluxes above forests: observations, methodological considerations and method comparisons. Environmental Pollution 152, 667–678.

Seibert, P., Beyrich, F., Gryning, S.E., Joffre, S., Rasmussen, A., Tercier, P., 2005. Review and intercomparison of operational methods for the determination of the mixing height. Atmospheric Environment 34, 1001–1027.

Sullivan, P.P., Moeng, C.-H., Stevens, B., Lenschow, D.H., Mayor, S.D., 1998. Structure of the entrainment zone capping the convective atmospheric boundary layer. Journal of the Atmospheric Sciences 55, 3042–3064.

Tomlin, A.S., Smalley, R.J., Tate, J.E., Barlow, J.F., Belcher, S.E., Arnold, S.J., Dobre, A., Robins, A., 2009. A field study of factors influencing the concentrations of a traffic-related pollutant in the vicinity of a complex urban junction. Atmospheric Environment 43, 5027–5037.

Vakeva, M., Hameri, K., Kulmala, M., Lahdes, R., Ruuskanen, J., Laitinen, T., 1999. Street level versus rooftop concentrations of submicron aerosol particles and gaseous pollutants in an urban street canyon. Atmospheric Environment 33, 1385–1397.

Vardoulakis, S., Fisher, B.E.A., Pericleous, K., Gonzalez-Flesca, N., 2003. Modelling air quality in street canyons: a review. Atmospheric Environment 37, 155–182.

Wilson, W.E., Mar, T., Kegler, S., 2001. Choice of indicator determines the significance and risk obtained from the statistical association between fine particulate matter mass and cardiovascular mortality. Journal of Aerosol Science 32 (Suppl. 1), S1081–S1082.

Weber, S., Kuttler, W., Weber, K., 2006. Flow characteristics and particle mass and number concentration variability within a busy urban street canyon. Atmospheric Environment 40, 7565–7578.

Wehner, B., Wiedensohler, A., 2003. Long term measurements of submicrometer urban aerosols: statistical analysis for correlations with meteorological conditions and trace gases. Atmospheric Chemistry and Physics 3, 867–879.

Wilson, J.D., Kingham, S., Pearce, J., Sturman, A., 2005. A review of intraurban variations in particulate air pollution: implications for epidemiological research. Atmospheric Environment 39, 6444–6462.

Zeger, S.L., Dominici, F., McDermott, A., Samet, J.M., 2008. Mortality in the Medicare population and chronic exposure to fine particulate air pollution in urban centers (2000–2005). Environ Health Perspectives 116 (12), 1614–1619.