Ultrafine particles in four European urban environments: first results of a continuous long-term monitoring network

J. Hofman[1]*, J. Staelens[1], R. Cordell[2], C. Stroobants[1], S.M.L. Hama[2,3], K.P. Wyche[4], G.P.A. Kos[5], C. Matheeussen[1], S. Van Der Zee[6], K.L. Smallbone[4], E.P. Weijers[5], P.S. Monks[2], E. Roekens[1]

[1]*Flanders Environment Agency (VMM), Department Air, Environment and Communication, Belgium*
[2]*University of Leicester, Department of Chemistry, United Kingdom*
[3]*Department of Chemistry, School of Science, University of Sulaimani, Kurdistan Region, Sulaimani, Iraq*
[4]*Air Environment Research, University of Brighton, School of Environment and Technology, United Kingdom*
[5]*Energy research Centre of the Netherlands (ECN), Environment & Energy Engineering, the Netherlands*
[6]*Public Health Service of Amsterdam, Department of Air Quality, the Netherlands*

**Present address corresponding author:*
University of Antwerp
Department of Bioscience Engineering
Groenenborgerlaan 171
2020 Antwerp
Belgium

Abstract

To gain a better understanding on the spatiotemporal variation of ultrafine particles (UFPs) in urban environments, this study reports on the first results of a long-term UFP monitoring network, set up in Amsterdam (NL), Antwerp (BE), Leicester (UK) and London (UK). Total number concentrations and size distributions were assessed during 1-2 years at four urban background sites, supplemented with a mobile trailer for co-location monitoring and additional short-term sites. Intra- and interurban spatiotemporal UFP variation, associations with commonly-monitored pollutants (PM, NO_x and BC) and impacts of wind fields were evaluated. Although comparable size distributions were observed between the four cities, source-related differences were demonstrated within specific particle size classes. Total and size-resolved particle number concentrations showed clear traffic-related temporal variation, confirming road traffic as the major UFP contributor in urban environments. Associations with typical traffic-related pollutants (BC and NO_x) were obtained for all monitoring stations, except for Amsterdam, where Schiphol airport was a significant contributor to the measured UFPs. The temporal variation in particle number concentration correlated fairly weakly between the four cities (r_s = 0.28-0.50, COD = 0.28-0.37), yet improved significantly inside individual cities (r_s = 0.59-0.77). Nevertheless, considerable differences were still obtained in terms of particle numbers (20-38% for total particle numbers and up to 49% for size-resolved particle numbers), confirming the importance of local source contributions and the need for careful consideration when allocating UFP monitoring stations in heterogeneous urban environments.

1. Introduction

Atmospheric aerosols, ranging from several nanometers to approximately 100 micrometers in diameter, are composed of primary particles emitted from both anthropogenic activities and natural sources, and secondary particles formed by gas-to-particle conversion processes including nucleation and condensation (Donaldson et al., 2001; Querol, 2011; Viana et al., 2015). They are typically characterized by varying size modes, i.e. <10 nm (nucleation), 10-100 nm (Aitkin mode), 100 nm - 1 µm (accumulation mode) and coarse mode (>1 µm), providing information on the contributing

emission sources and attributing chemical and physical processes (Vu et al., 2015). Current air quality legislation focusses on monitoring, limiting and reducing mass concentrations of these airborne particles. However, recent toxicological and epidemiological research suggests that particle numbers may constitute better links to health endpoints than mass concentration (Donaldson et al., 2001; Harrison et al., 2000; Kelly and Fussell, 2012; Viana et al., 2015). In particular ultrafine particles (UFPs), consisting of aerosols smaller than 100 nm, have been shown to cause adverse health effects owing to their ability to penetrate deeply into the respiratory system and enter the bloodstream inducing inflammation, resulting in cardiovascular and respiratory conditions. In ambient air, ultrafine aerosols are dominant in terms of particle number (80-90% of all particles), but negligible in terms of particle mass, and are, therefore, inadequately quantified in current (mass-based) air quality monitoring networks. This especially holds true in urban areas, where concentrated local emissions sources and a complex urban topography are known to reduce pollutant dispersion. Consequently, there is a clear need for a thorough understanding of the spatiotemporal variation of UFPs.

There have been several short-term studies which have contributed to existing knowledge on the number and size distribution of specific UFP sources, and attributing formation and transformation processes of UFPs (Brines et al., 2015; Dall'Osto et al., 2013; González et al., 2011; Hudda et al., 2014; Keuken et al., 2015; Kozawa et al., 2012; Zhu et al., 2002). Studies reporting on long-term simultaneous UFP measurements at multiple sites are, however, scarce (Pey et al., 2008; von Bismarck-Osten et al., 2013). Nevertheless, such networks are vital to elucidate the complex relationship between local emission sources, meteorological processes, atmospheric transformation and the resulting aerosol number, size and distribution at sites with differing characteristics. This study reports on the first results of a novel North-West European UFP monitoring network, established in Amsterdam, Antwerp, Leicester and London, and was carried out as part of the Joint Air Quality Initiative (www.joaquin.eu), an INTERREG IVB funded European project, which aims at supporting health-oriented air quality policies in Europe. The main aims were to gain more insight in the spatiotemporal variation in UFP number concentration and size distribution and to assess the added value of UFP data compared to more commonly measured parameters such as black carbon (BC) and nitrogen oxides (NO_x).

2. Material and methods

2.1 Monitoring sites

An UFP monitoring network was set up in four NW European cities (*Figure 1*), consisting of four fixed monitoring sites at urban background locations in Amsterdam (the Netherlands; AD1), Antwerp (Belgium; AP1), Leicester (United Kingdom; LE1) and London[1] (United Kingdom; LO1). In addition to the fixed monitoring sites, a mobile monitoring unit was deployed for comparative UFP measurements collocated with all fixed monitoring sites (1M) and for additional UFP measurements at a second urban background site (2M) in Amsterdam (6.2 km from AD1), Antwerp (1.3 km from AP1) and Leicester (1.2 km from LE1). Hence, UFPs were measured at seven urban background locations across NW Europe (*Figure 1*).

The UFP measurements started in April, 2013, in Amsterdam and Antwerp, and later in Leicester (November 2013) and London (April 2014) owing to legislation issues. Results up to March, 2015, are discussed, hence the discussion covers a period of 1 to 2 years depending on the site considered. The measurements by the mobile monitoring unit were carried out during 2-4 weeks next to the fixed

stations and during 2-7 weeks at the additional urban background sites (AD2M, AP2M, LE2M) (Table 1).

[1]*Now moved to Brighton (United Kingdom)*

Table 1: Overview of the applied fixed and mobile unit monitoring sites of the UFP monitoring network

City	Code	Fixed/Mobile	Name	Nearest street	Coordinates		Monitoring period	
					Latitude	Longitude	Start	End
Amsterdam	AD1	Fixed	Vondelpark	Overtoom	52°21'35" N	4°51'59" E	01/04/2013	31/03/2015
	AD1M	Mobile	Vondelpark	Overtoom	52°21'35" N	4°51'59" E	17/04/2013	14/05/2013
	AD2M	Mobile	Nieuwendammerdijk	Nieuwendammerdijk	52°23'21" N	4°56'38" E	14/05/2013	30/05/2013
Antwerp	AP1	Fixed	Borgerhout	Plantin en Moretuslei	51°12'35" N	4°25'55" E	01/04/2013	31/03/2015
	AP1M	Mobile	Borgerhout	Plantin en Moretuslei	51°12'35" N	4°25'55" E	04/11/2013	19/11/2013
	AP2M	Mobile	Stadspark	Rubenslei	51°12'48" N	4°24'51" E	07/10/2013	04/11/2013
Leicester	LE1	Fixed	Leicester University	Welford Road	52°37'12" N	1°07'38" E	25/10/2013	31/03/2015
	LE1M	Mobile	Leicester University	Welford Road	52°37'12" N	1°07'38" E	04/03/2014	04/04/2014
	LE2M	Mobile	Brookfield	London Road	52°37'15" N	1°06'32" E	05/04/2014	29/05/2014
London	LO1	Fixed	Eltham	Bexley Road	51°27'09" N	0°04'14" E	21/04/2014	31/03/2015
	LO1M	Mobile	Eltham	Bexley Road	51°27'09" N	0°04'14" E	02/06/2014	30/06/2014

Figure 1: Overview of the UFP monitoring network: four fixed urban background sites in Amsterdam (AD1; NL), Antwerp (AP1; BE), Leicester (LE1; UK) and London (LO1; UK) and the mobile monitoring unit for additional UFP measurements at a second urban background site in three cities (AD2M, AP2M and LE2M).

2.2 Instrumentation

2.2.1 Air quality data

Several commercially available UFP instruments were evaluated via a comprehensive literature review and laboratory test, in order to choose the most appropriate instrumentation and methodology for particle number and size distribution measurements under continuous monitoring network conditions. Based on this evaluation, three instruments were selected for application in the UFP monitoring network (*Table 2*).

Table 2: Specifications of the employed UFP instrumentation

Name	*Company/type*	*Lower size (nm)*	*Upper size (nm)*	*UFP size classes*	*Sample time (min)*	*Radioactive source*	*Condesation fluid*
EPC	TSI 3783	7	1000	1	1	-	Water
UFPM	TSI 3031	20	500	6	10	-	-
SMPS	Grimm 5420+C L-DMA	10	1000	45	10	^{85}Kr (185 Mbq)	Butanol

Total UFP number concentrations (# cm^{-3}) were obtained by means of a water-based Environmental Particle Counter (EPC) at each monitoring station. After initial tests, the high-flow inlet mode (3 l min^{-1}) was applied to minimize particle losses. Size-resolved particle number concentrations (# cm^{-3}) were obtained using two different instruments (UFPM and SMPS) owing to legislation issues with the radioactive source (^{85}Kr) at the UK sites. In Amsterdam (AD1) and Antwerp (AP1), particle number concentrations in 45 different size classes were obtained by a scanning mobility particle sizer (SMPS). In Leicester (LE1) and London (LO1), UFPs were quantified in six size classes (20-30, 30-50, 50-70, 70-100, 100-200 and >200 nm), using an UFPM (Table 2). In brief, the operating principle of the SMPS comprises radioactive (^{85}Kr) charging of particles, followed by size segregation based on particle electrical mobility using a differential mobility analyser (L-DMA) and particle counting by means of a butanol-based condensation particle counter (CPC). The UFPM principle of operation is based on electrical diffusion charging of the particles, size segregation by means of a DMA, followed by aerosol detection using a Faraday cup electrometer.

A Multi-Angle Absorption Photometer (MAAP 5021, Thermo Scientific) was installed in all monitoring stations to measure ambient black carbon (BC) concentrations (μg m^{-3}). In addition to the UFP and BC instruments in the fixed monitoring stations, continuous air quality monitors were already available for NO_x (Thermo 42i in AP1, LE1 and LO1 and a API 200A in AD1), PM_{10} (BAM1020 in AD1, ESM FH62 I-R and FIDAS 200 in AP1 and TEOM-FDMS in LO1) and $PM_{2.5}$ (BAM1020 in AD1, ESM FH62 I-R and FIDAS 200 in AP1 and TEOM-FDMS in LE1 and LO1). The mobile monitoring unit was equipped with all UFP instruments (EPC, UFPM, SMPS) and a MAAP 5012 for atmospheric BC measurements. For the EPC and UFPM instruments an Environmental Sampling System (ESS; TSI 3031200) was used with a PM_{10} inlet, sharp-cut PM_1 cyclone and Nafion dryer. The EPC in AD1 and AP1 were individually connected to an ESS. In LE1, LO1 and the trailer, two instruments (EPC and UFPM) were connected to one ESS. The SMPS devices had an individual Grimm sampling system with TSP inlet and Nafion dryer. Standard operating procedures were created for the applied instrumentation to ensure that comparable monitoring data was collected at the seven locations (monitoring artefacts, e.g. inlet systems, maintenance frequency etc.).

Before the instruments were installed at the monitoring sites, they were intercompared in an initial co-location monitoring campaign from December, 2012, to January, 2013, at an urban background

location in Antwerp (Frijns et al., 2013a). All EPCs and SMPSs were strongly correlated and differed by less than 10% (except for the LE1 EPC; 13%, probably due to the sampling setup which was changed following the colocation trial). The total number concentration, quantified by the EPC, was approximately 20% higher compared to the SMPS and 24% higher compared to the UFPM. More details on the instrument comparisons can be found in the report by (Frijns et al., 2013a). After installing the instruments at their monitoring locations, the mobile monitoring unit conducted measurements adjacent to each monitoring site to evaluate the comparability of the instruments and reliability of the conducted measurements. Results of the mobile monitopring unit comparison can be found in the final Joaquin reporting (in prep).

2.2.2 Meteorological data

Meteorological data of ambient air temperature (°C), relative humidity (%), atmospheric pressure (Pa), wind direction (°) and speed (m s^{-1}) were obtained for each monitoring site. Meteorological parameters (e.g. wind) can be altered significantly at the local scale due to the urban canopy (e.g. building height, street orientation,…). Therefore, regional meteorological data were collected to enable evaluation of larger-scale air mass transport processes. Regional meteorology was measured at 9 km from AD1 (Schiphol airport), 6 km from AP1 (Luchtbal monitoring station of the Flanders Environment Agency, VMM), 5 km from LE1 (Groby Road monitoring station) and 14 km from LO1 (Barking and Dagenham – Rush Green monitoring station).

2.3 Data validation and treatment

The raw 10 minute-data were validated by screening for irregularities and removing data collected during instrument errors and maintenance periods. All validated data were subsequently aggregated to 30 minute intervals. The retain threshold in further data averaging was 75% availability at the half-hourly level. For comparison purposes between the considered monitoring sites, size-resolved UFP concentrations, obtained by the SMPS (45 size classes), were aggregated to the UFPM size classes: 10-20, 20-30, 30-50, 50-70, 70-100 and 100-200 nm.

Boxplots, single linear regression plots, coefficients of divergence (COD) and Spearman Rank (r_s) correlations were applied to compare monitoring sites, time periods and pollutants. Potential effects of wind speed and direction were evaluated using pollution roses and polar plots. All statistical analyses were performed using the statistical software package R (R Development Core Team, 2015), more specifically in the *openair* package (Carslaw and Ropkins, 2015, 2012).

3. Results and Discussion

3.1 Data exploration

The 30 minute air quality and meteorological data were collected for the entire sampling period, from April, 2013, to March, 2015. Taking into account the later start of the UFP measurements in Leicester and London (Table 1), data coverage at the 30 minute was 96% for BC, 79% for total particle number concentrations (TNC) and 83% for size-resolved particle number concentrations (PNC). This is comparable but generally lower than for the commonly monitored pollutants NO_2 (89%), PM_{10} (94%) and $PM_{2.5}$ (81%). Boxplots of the log-transformed PM, NO_2, BC and TNC concentrations showed comparable variability between the various monitoring stations. Nevertheless, the site in Antwerp showed higher overall concentrations of the typical traffic-related pollutants (NO_2, BC and TNC),

compared to the other sites (*Figure 2*). This can be explained by its proximity (30 m) to a traffic-intensive access road into Antwerp (*Plantin en Moretuslei*). In February and October 2013, the mean traffic volume was 32000 vehicles on weekdays and 23500 vehicles in the weekend; or a time-weighted average of 29500 vehicles/day (VMM, 2014).

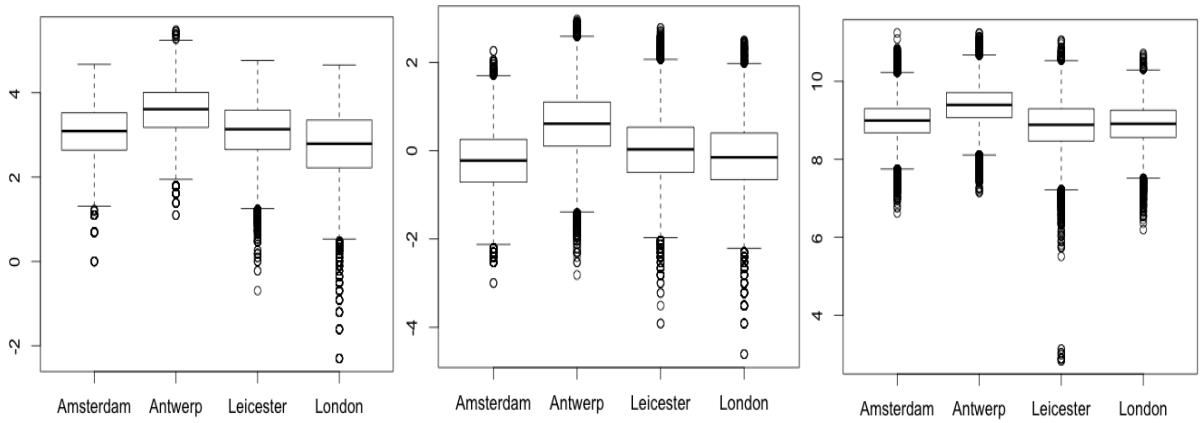


Figure 2: Boxplots of the obtained log-transformed half-hourly NO_2 (left), BC (middle) and TNC (right) concentrations at the individual monitoring sites.

Looking at the range of the quantified total and size-resolved PNC (*Table 3*), comparable UFP variability was found at the monitoring sites, with the highest PNC observed in Antwerp. For all monitoring sites, the highest PNC were obtained in the smallest particle size class (10-20 nm), consecutively followed by the 30-50, 20-30, 50-70, 70-100 and 100-200 nm size classes. In Leicester and London, the 10-20 nm size class was not quantified due to the size range restrictions of the applied UFPM (see *Table 2*). Nevertheless, comparable behaviour of the 10-20 nm size class was observed from co-located SMPS measurements during the 2-4 week instrument comparison conducted by the co-located mobile monitoring unit (Joaquin, 2015).

Table 3: Range (25% quartile, mean, 75% quartile and maximum) of the half-hourly PM, NOx, BC, total (TNC) and size-resolved (PNC) particle number concentrations, measured at the fixed monitoring sites in Amsterdam (AD1), Antwerp (AP1), Leicester (LE1) and London (LO1)

	Amsterdam (AD1)	*Antwerp (AP1)*	*Leicester (LE1)*	*London (LO1)*		*Amsterdam (AD1)*	*Antwerp (AP1)*	*Leicester (LE1)*	*London (LO1)*
PM_{10} (µg m^{-3})					PNC 10-20 nm (# cm^{-3})				
25% quartile	12.24	15.00	-	11.30	25% quartile	1125	1327	-	-
mean	20.64	25.99	-	18.64	mean	2592	2468	-	-
75% quartile	25.21	32.50	-	22.50	75% quartile	2956	3093	-	-
max	227.50	176.25	-	122.50	max	56575	35412	-	-
$PM_{2.5}$ (µg m^{-3})					PNC 20-30 nm (# cm-3)				
25% quartile	6.82	7.00	6.70	6.10	25% quartile	805	974	755	475
mean	14.24	16.17	13.47	13.00	mean	1552	1709	1541	1007
75% quartile	17.66	20.47	16.70	15.90	75% quartile	1773	2112	2001	1191
max	225.30	145.00	181.00	90.40	max	39199	19634	13795	29072
NO_2 (µg m^{-3})					PNC 30-50 nm (# cm-3)				
25% quartile	14.00	24.00	14.20	9.20	25% quartile	1031	1278	891	811
mean	25.49	41.37	27.13	20.63	mean	1773	2195	1774	1539
75% quartile	34.00	55.00	36.20	28.60	75% quartile	2163	2704	2227	1946
max	107.00	242.00	117.80	105.70	max	19756	26669	16641	22534
NO (µg m^{-3})					PNC 50-70 nm (# cm-3)				
25% quartile	0.40	2.00	1.80	1.30	25% quartile	537	717	594	426
mean	4.89	17.56	11.07	6.60	mean	950	1267	1247	809
75% quartile	4.00	18.00	10.60	4.90	75% quartile	1215	1598	1539	1042
max	230.03	784.00	540.00	321.10	max	8907	15387	14614	8959
BC (µg m^{-3})					PNC 70-100 nm (# cm-3)				
25% quartile	0.49	1.11	0.61	0.52	25% quartile	362	553	504	400
mean	1.01	2.36	1.40	1.22	mean	759	1063	1112	776
75% quartile	1.29	3.00	1.70	1.49	75% quartile	1026	1382	1363	1012
max	9.56	19.52	16.05	12.13	max	5546	5765	17444	10074
TNC (# cm^{-3})					PNC 100-200 nm (# cm-3)				
25% quartile	5889	8713	4760	5230	25% quartile	363	604	447	319
mean	9070	13481	8623	8353	mean	807	1182	1010	711
75% quartile	10952	16538	10916	10506	75% quartile	1069	1531	1233	936
max	76549	76170	63481	45155	max	20116	11903	19702	12707

3.2 Relationship with commonly-monitored pollutants

To evaluate potential relationships between UFPs and more commonly monitored atmospheric pollutants, 30 minute and daily-averaged TNC was plotted against PM_{10}, $PM_{2.5}$, NO_2, NO and BC concentrations per site. The TNC was linearly related with BC (*Figure 3*), NO_2 (not shown) and NO (not shown), which confirms vehicle engines as an important source of UFPs at the studied sites, as reported earlier (Goel and Kumar, 2015; Kumar et al., 2014; Mishra et al., 2012; Querol, 2011).

Nevertheless, for the Amsterdam site, relationships between these typical traffic-related pollutants and TNC were significantly weakened. Therefore, traffic may not be the dominant UFP source at this particular monitoring location. The presence of the low emission zone (Panteliadis et al., 2014) and/or contributions from other UFP sources might explain this lack of correlation between traffic-related pollutants and UFP number concentration in Amsterdam.

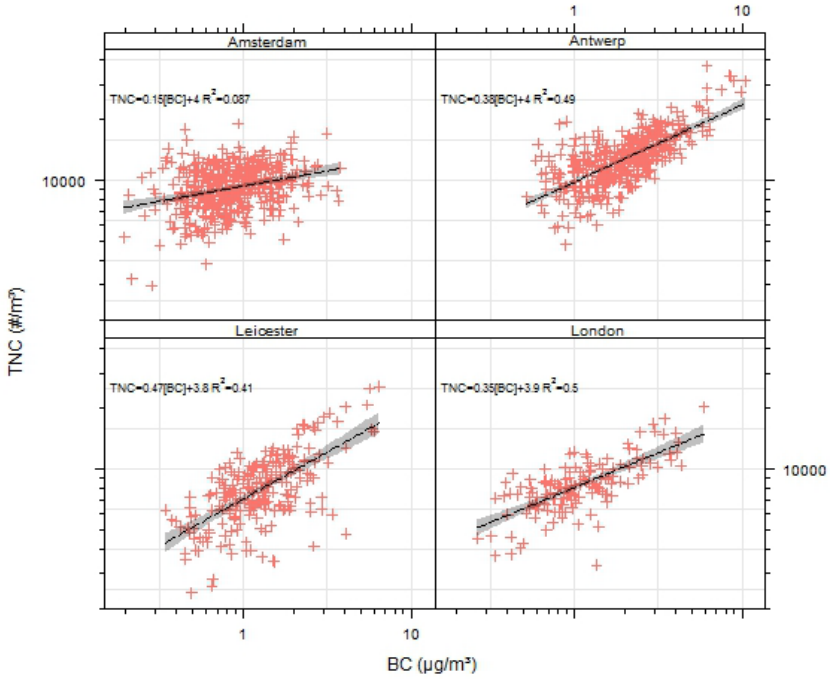


Figure 3: Regression plots of daily-averaged total particle number concentration (#/cm³) and BC (µg/m³) at the fixed sites (AD1, AP1, LE1 and LO1). Note the logarithmic scales for both pollutants.

The relationships observed between the atmospheric pollutants seemed to exhibit a seasonal variation (not shown). For Antwerp, the highest correlation obtained between BC and TNC was during the winter season (R^2 = 0.64). The relationship was weakest during the summer season (June, July, August), which may suggest a higher contribution of non-traffic emitted UFPs, e.g. originating from new particle formation.

3.3 Temporal variation in TNC

Temporal variation plots of hourly-, daily- and monthly-averaged TNC confirmed that the site in Antwerp experienced higher TNC, compared to Amsterdam, Leicester and London (*Figure 4*). A typical traffic-related diurnal variation was observed throughout the day, with distinct morning and evening peaks coinciding with traffic rush hours. During the weekends, the peaks were less pronounced and almost negligible for the morning rush hour, which seems to confirm road traffic as

the main UFP attributor in urban environments. This was also confirmed when compared to the temporal variation of BC (not shown Appendix 1?), which can be considered as a typical traffic-related pollutant. Similar diurnal variations, with distinct morning and evening peaks, and decreased concentrations during the weekend were identified. For all monitoring sites, highest monthly-averaged TNC were obtained during winter months (September-March). This is likely due to meteorological conditions (e.g. temperature and mixing layer height) favouring higher atmospheric UFP concentrations, as reported before by Mishra et al. (2012), Pey et al. (2008) and von Bismarck-Osten et al. (2013).

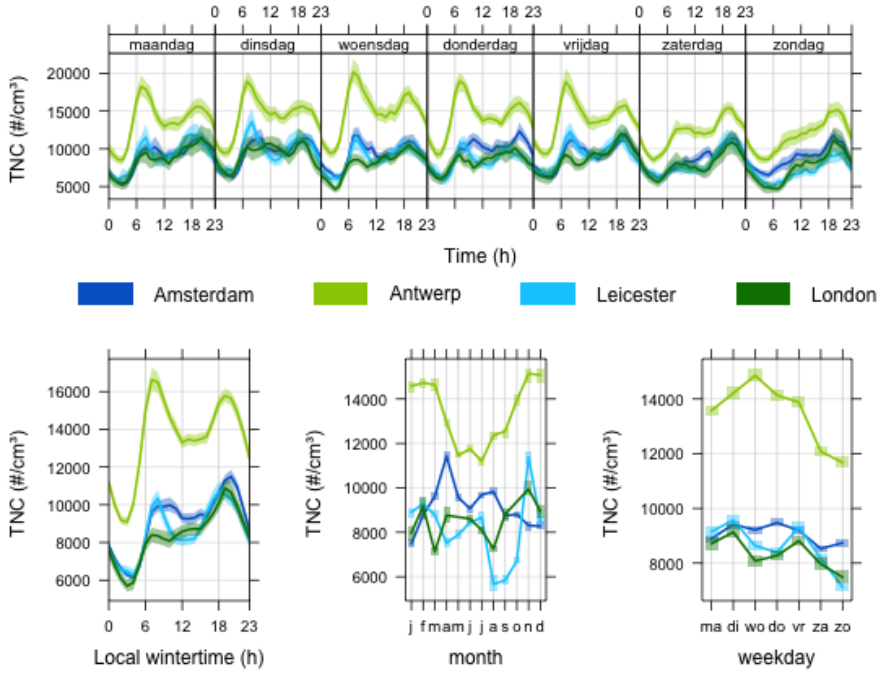


Figure 4: Temporal variation of total particle number concentration (TNC; # cm^{-3}) at the four fixed monitoring sites (AD1, AP1, LE1 and LO1) at three different time scales (hourly, daily and monthly averages). The coloured zone represents the 95% confidence interval.

For the hourly-averaged diurnal UFP variation per particle size class (*Figure 5*), comparable findings as for the TNC were observed, with a more or less constant ratio of the individual size classes, indicating a fairly stable UFP size distribution throughout time (also observed for the daily- and monthly-averaged PNC). Nevertheless, temporal differences were obeserved for the 10-20 nm particle size class, which was only quantified in Amsterdam and Antwerp. For Amsterdam, a much higher relative contribution of the 10-20 nm class with respect to the other particle size classes was found compared to Antwerp (*Figure 5*). Moreover, a constant contribution (>3000 particles cm^{-3}) was observed throughout the day (7:00-20:00h), while in Antwerp, the 10-20 nm sized particles followed the morning and evening rush hour peaks. Also during the weekends, an average constant contribution of 10-20 nm sized particles was observed, while the PNC of all other size classes are observed to decrease considerably (not shown). These data, therefore, suggest a non-traffic related input of mainly smaller-sized particles in Amsterdam. This UFP source seems to persist throughout the weekend, with the 10-20 nm size channel exhibiting a diurnal variation that is comparable to that observed during the working week. There was no clear decrease in the average PNC during the weekends, nor was there a seasonal influence.

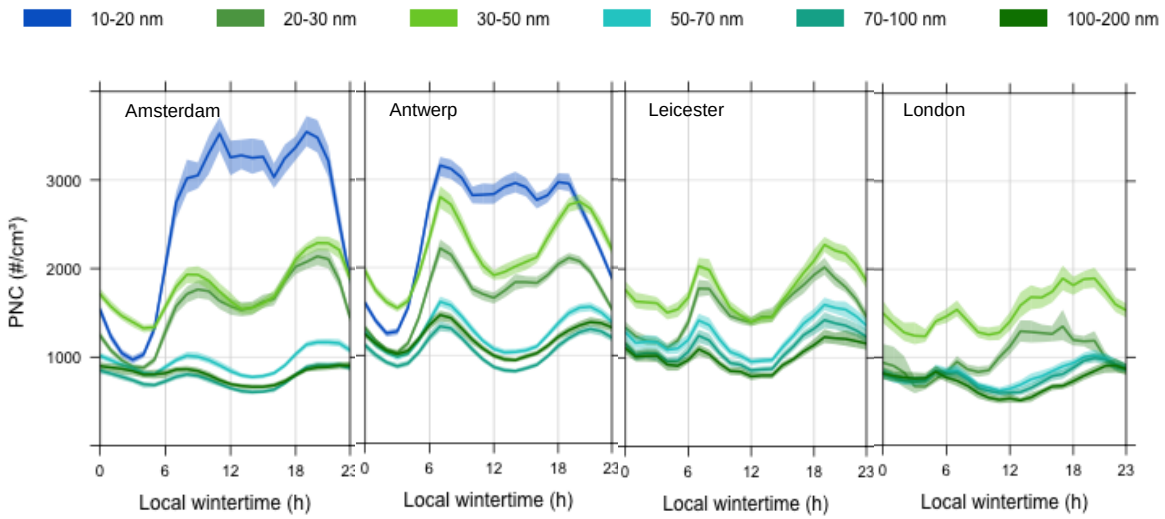


Figure 5: Temporal variation of the hourly-averaged particle number concentration (PNC; # cm^{-3}) within the individual particle size classes (10-20 nm, 20-30 nm, 30-50 nm, 50-70 nm, 70-100 and 100-200 nm) at the four fixed monitoring sites (AD1, AP1, LE1 and LO1).

In Antwerp, the hourly-averaged 10-20 nm sized particles showed a small midday-peak, which was not observed for the other particle size classes (only to a limited extent in the 30-50 nm size class). This observation resembles at new photochemical particle formation (nucleation) events in urban areas as described in former studies (Brines et al., 2015; Kulmala and Kerminen, 2008; Pey et al., 2008; Querol, 2011; Wang et al., 2014). In Brisbane, Barcelona, Madrid, Los Angeles and Rome, Brines et al. (2015) observed photochemical nucleation events at midday, under high temperature, solar radiation and ozone levels, which lasted for 2 to 4 hours. We, therefore, tested for this hypothesis by selecting for midday-early afternoon hours (11:00-15:00 local wintertime) and plotting the measured 10-20 nm PNC (#/cm^3) against the solar radiation (W/m^2), temperature (T, °C) and ozone (O_3, µg/m^3). However, no relation could be observed (R^2 < 0.01 for radiation, T and O_3). Nevertheless, when plotting the hourly-averaged detailed SMPS size distributions throughout the day, 10-20 nm particle bursts starting around noon (13:00-14:00h) can be observed on weekdays and during the weekends. These particle bursts last for approximately 2-4 hours during which a modest growth in particle diameter can be observed of up to 40 nm, which is eventually suppressed by the condensation sink of the evening rush hour (*Figure 6*). Although less pronounced, these events are very similar to the nucleation events observed in Mediterranean areas (Brines et al., 2015; Pey et al., 2008) and confirm the existence of new photochemical particle formation events in Antwerp. Noon particle bursts within the 10-20 nm particle size class were also observed in Amsterdam, although less obvious due to the continuous 10-20 nm contribution throughout the day and weekend. No clear nucleation episodes could be identified for Leicester, however a number of particle growth events, consistent with new particle formation, were observed in London. Owing to the size resolution of the UFPM (only > 20 nm), these growth events were observed to begin in the (lowest) 20 – 30 nm channel. Further details regarding these measurements made in London will be discussed in detail in a separate piece of work (Wyche et al., 2016, in preparation).

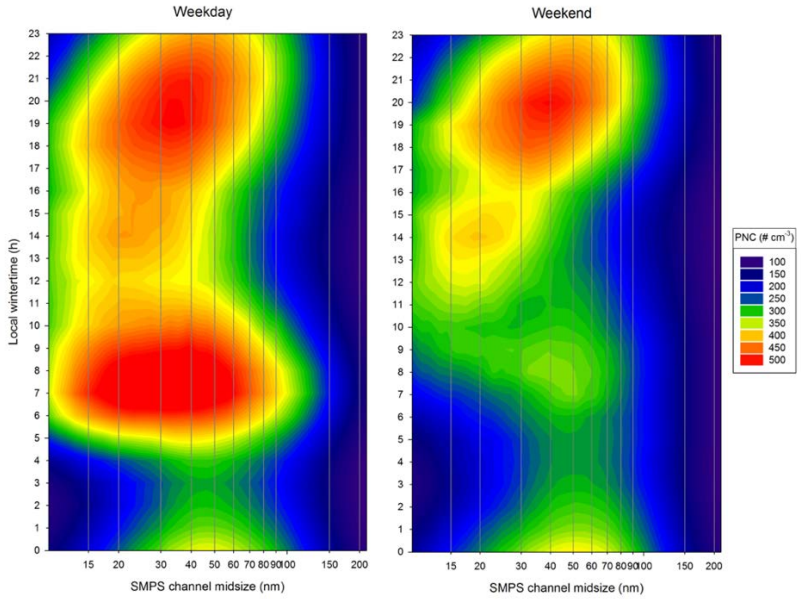


Figure 6: Hourly-averaged PNC (# cm^{-3}), obtained by the SMPS (selected for 33 size classes below 200 nm) during weekdays (left) and weekends (right) at the Antwerp monitoring site (AP1).

3.4 Spatial variation

3.4.1 Inter-urban

The average UFP size distributions within the aggregated particle size classes (*Figure 7*) were fairly similar between the considered monitoring locations. Nevertheless, Antwerp seems to have a proportionally higher contribution of 30-50 nm sized particles, while the 10-20 nm size range is proportionally higher in Amsterdam. When normalized for size bin width (dN (dlog Dp)$^{-1}$), highest particle numbers are obtained near 30-50nm, except for Amstedam (20 nm). The total particle number concentration was significantly higher in Antwerp, compared to the other monitoring sites (*Figure 7*). This can be explained by considering the proximity (30 m) of the monitoring site to a very busy access road of Antwerp (*Plantin en Moretuslei*). All other monitoring sites are located further away from road traffic (*Figure 1*) and their nearest roads have lower traffic volumes.

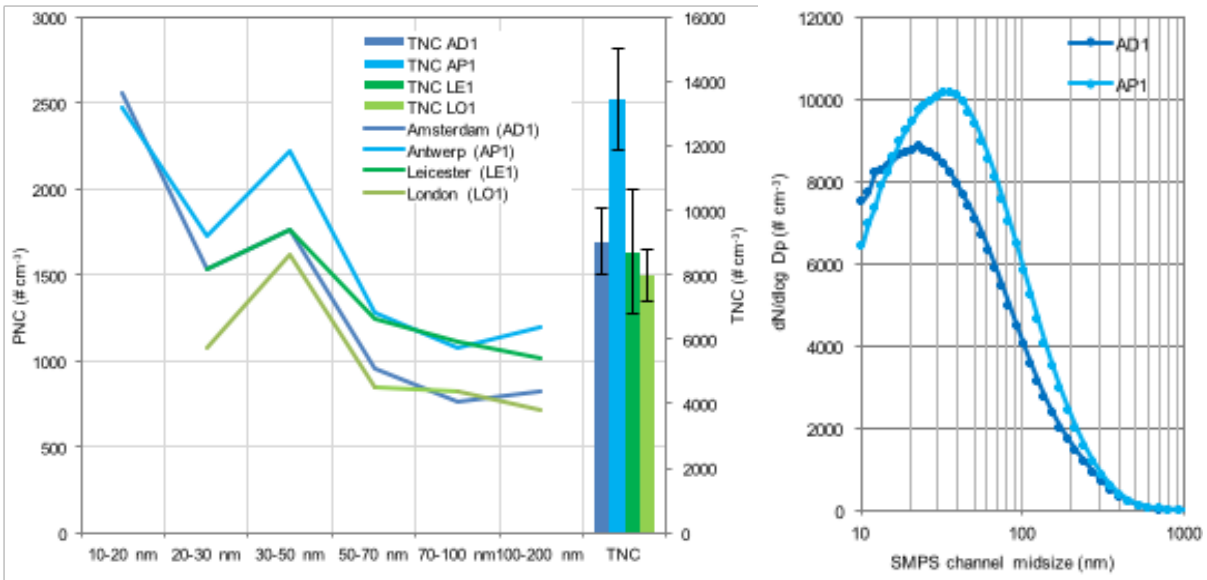


Figure 7: Average size-resolved (PNC; lines) and total (TNC; bars) particle number concentrations for the fixed monitoring locations in Amsterdam, Antwerp, Leicester and London (left) and the full SMPS size distributions with 45 size classes ($dN/dlogD_p$), obtained in Amsterdam and Antwerp (right).

The spatial TNC variation was evaluated by calculating the coefficients of divergence (COD) and Spearman rank correlation coefficients (r_s) between data pairs of the considered monitoring sites (*Table 4*). Most variation in TNC is observed between the monitoring sites in Antwerp and Leicester (COD = 0.37, r_s = 0.30), while the best agreement in TNC was found between Leicester and London (COD = 0.28, r_s = 0.50). This is not surprising, as London and Leicester are most closely located to one another and are located under the same national governance. Overall, correlations are fairly low (≤ 0.5) indicating that TNC is not related at the regional level of NW Europe and that much of the variation in TNC is therefore owing to local factors.

Table 4: Coefficients of determination (COD, left) and Spearman rank correlations (r_s, right) of the half-hourly total particle number concentration (TNC) between the respective monitoring sites.

COD TNC				
	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.32	0.37	0.33
Amsterdam	0.32	0.00	0.32	0.29
Leicester	0.37	0.32	0.00	0.28
London	0.33	0.29	0.28	0.00

Spearman rank (r_s) TNC				
	Antwerp	Amsterdam	Leicester	London
Antwerp	1	0.37	0.30	0.38
Amsterdam	0.37	1	0.31	0.28
Leicester	0.30	0.31	1	0.50
London	0.38	0.28	0.50	1

From the COD and correlation coefficients of the individual size classes in Appendix 3, an increased association (smaller COD and larger correlation) is obtained with increasing particle size. Therefore, larger particles tend to be more uniform, which may indicate the regional nature of these aerosols. Long-range transported aerosols comprise mostly accumulation mode particles, with the major number peak mode around 100-200 nm (Vu et al., 2015). (Krudysz et al., 2009) previously found an inverse relationship between particle size and CODs for 13 different monitoring locations within 350 m - 11 km of each other within the city of Los Angeles.

3.4.2 Intra-urban

To evaluate the spatial variation in total UFP numbers (TNC) within the investigated urban environments, a second urban background location (2T) was sampled by means of the mobile monitoring unit in Amsterdam, Antwerp and Leicester (*Table 1*). The raw TNC concentrations were averaged to hourly values and the hourly TNC concentrations measured by the fixed monitoring sites (1T) in *Figure 8*.

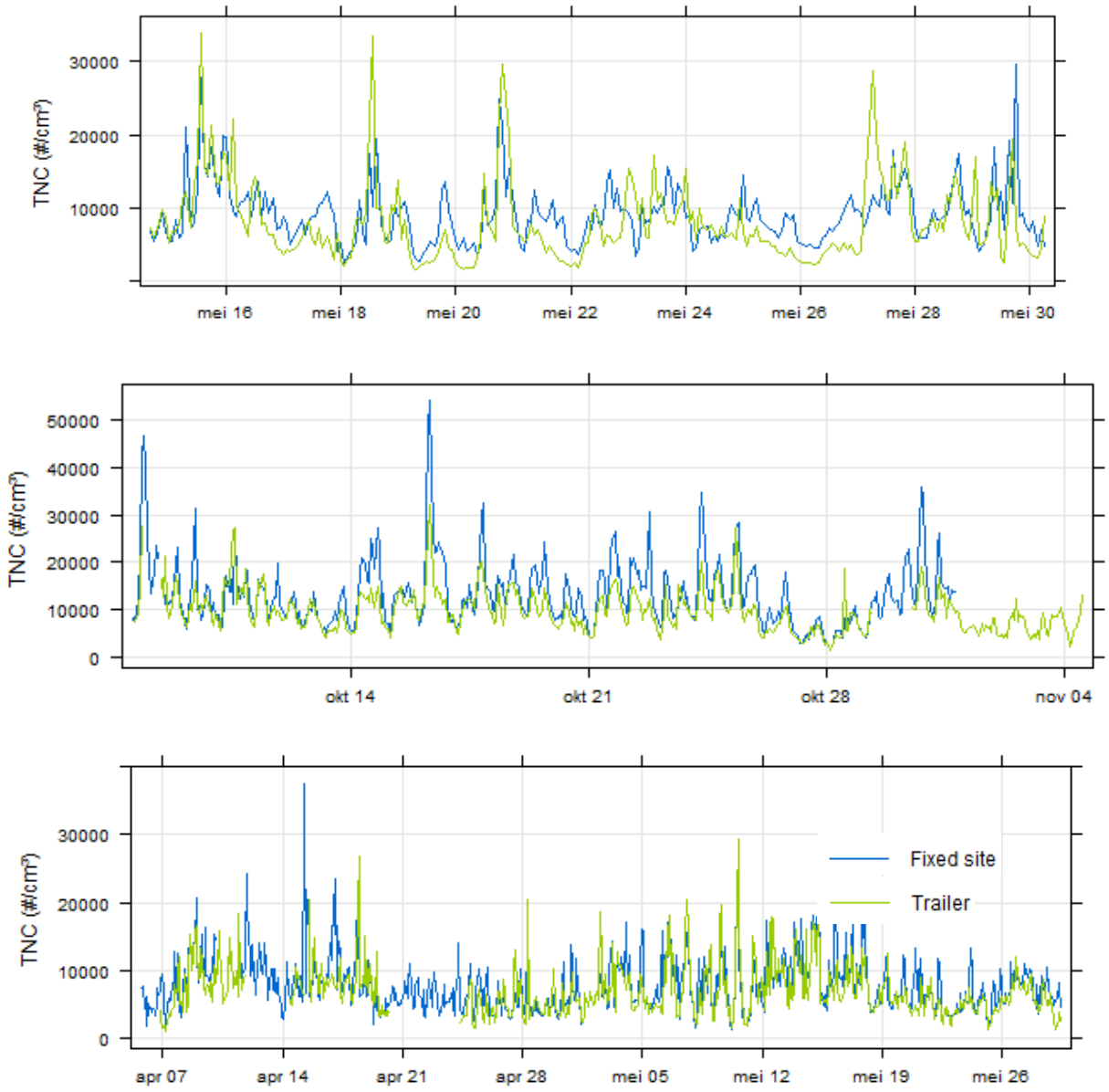


Figure 8: Temporal variation of the hourly-averaged total particle number concentration (# cm^{-3}) at the fixed and mobile unit locations in Amsterdam (top), Antwerp (middle) and Leicester (bottom).

The temporal variation plots (*Figure 8*) show that the TNC concentrations at the fixed and mobile monitoring unit locations covary in time. In particular for Antwerp and Leicester, the covariance between the two intra-urban locations seems good, while for Amsterdam some deviations between the two monitoring locations is observed. The temporal UFP variation seems to consist of two levels. First, there is a (slowly changing) base level which behaves roughly similar in time and magnitude at both paired sites. In particular, this is the case in Antwerp and Leicester, while in Amsterdam there is a small difference of roughly 3000 #/cm^3 between the sites. Looking at the individual particle size classes, it can be seen that this effect is predominantly observed in the 10-20 nm size class, which may be influenced by the different distances of teh fixed and mobile sites, respectively, to Schiphol airport. In addition to this base level, part of the fast variation is observed at both sites per city. A clear example is seen in the time series for Antwerp: the peaks at the Stadspark location (AP2) usually occur simultaneously at Borgerhout (AP1) but have a different magnitude. This is also found at the Leicester sites, and to a lesser extent, at the Amsterdam sites. This could be regarded as an

overall urban contribution mostly originating from traffic emissions following a similar behaviour in time but differing in quantity depending on the distance to these emissions source. Apart from these contributions, certain local effects were noted affecting one site but not the other, as can be seen in Amsterdam, which is likely due to a differing influence of a non-traffic source.

In addition to the time series plots, coefficients of divergence (COD) and Spearman Rank correlations (R_s) were calculated for the total number concentration (TNC) between the fixed and mobile monitoring unit locations in Amsterdam, Antwerp and Leicester. As already suggested by the time series plots, the highest association (lowest COD and highest r_s) is obtained for Antwerp (COD = 0.16 and r_s = 0.85), followed by Leicester (COD = 0.18 and r_s = 0.77) and Amsterdam (COD = 0.25 and r_s = 0.59).

Nevertheless, based on the average size distributions at the paired sites per city (*Figure 9*), it can be concluded that large proportional differences in number concentration are observed, depending on the particle size class considered. On average, the largest intra-urban variation in TNC was observed in Antwerp (38%), followed by Amsterdam (24%) and Leicester (20%). For Amsterdam, the 10-20 nm particle number concentration was 48% lower at the mobile unit location (AD2T, Nieuwendammerdijk), compared to the fixed monitoring station (AD1, Vondelpark). For Antwerp, the strongest difference in size distributions was observed, with up to 49% lower particle numbers for AP2T in the 100-200 nm size range. This is not surprising, as the mobile unit location was within an urban park (*Stadspark*), while the fixed monitoing site is located 30 m from a very busy access road (*Plantin en Moretuslei*). In Leicester, the largest difference was observed in the 70-100 nm size range, with 30% lower particle number concentrations at the mobile unit location (LE2T, Brookfield), compared to the fixed monitoring site (LE1, Leicester University).

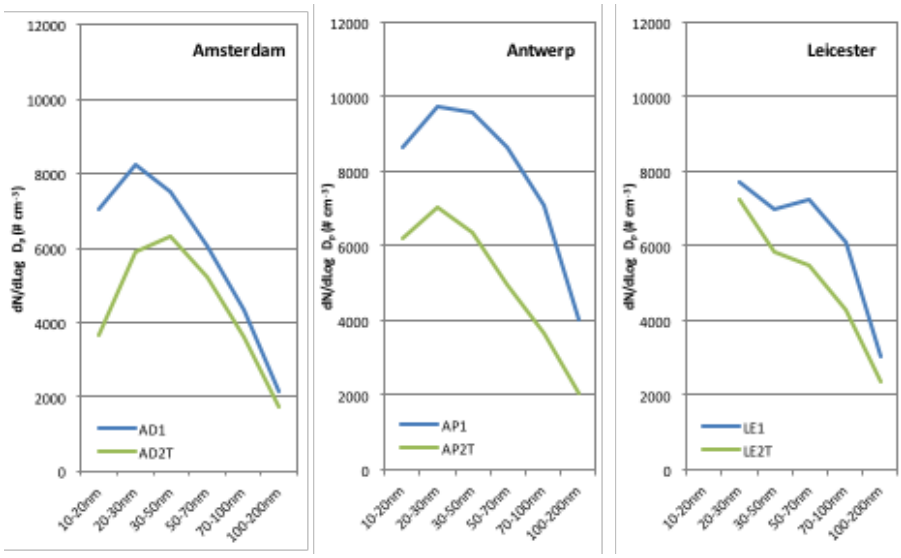


Figure 9: Average size-resolved PNC (dN (dlog D_p)$^{-1}$) at the fixed (1) and mobile unit (2T) locations in Amsterdam (left), Antwerp (middle) and Leicester (right).

Although the UFP number concentrations covary in time at the monitored locations, considerable proportional differences in size-resolved number concentrations are obtained between the individual intra-urban sites, influenced by their proximity to urban UFP sources. This implies that the location of the UFP monitoring station is of primordial importance when evaluating citizen's exposure to UFP in

urban environments. In epidemiological studies, UFP data from a single monitoring site is generally used as a measure of population exposure in a wider region. One reason for this is the lack of sufficient data at other sites, which may potentially result in exposure misclassification. While the spatial PM_{10} mass variation (or related particle mass parameters) is known to be quite low over an urban region, our results show that this is not the case for particle numbers.

3.5 Influence of wind field on measured UFP concentrations

All the monitoring sites in this study are classified as urban background stations. In order to assess the influence of local sources on the measured UFP concentrations, the potential effect of the experienced wind field on the total and size-resolved UFP number concentrations was evaluated. In former studies, wind direction and speed have namely shown to be dominant influencing factors in the spatial variability of UFP number concentrations (Keuken et al., 2015; Kozawa et al., 2012; von Bismarck-Osten et al., 2013). From the wind roses shown in *Figure 10*, it is clear that the main wind direction in Amsterdam, Antwerp and London is from the southwest. Average regional wind speeds over the entire sampling period were 4.8 m s^{-1} for Amsterdam, 4.1 m s^{-1} for Antwerp, 3.0 m s^{-1} for Leicester and 3.8 m s^{-1} for London.

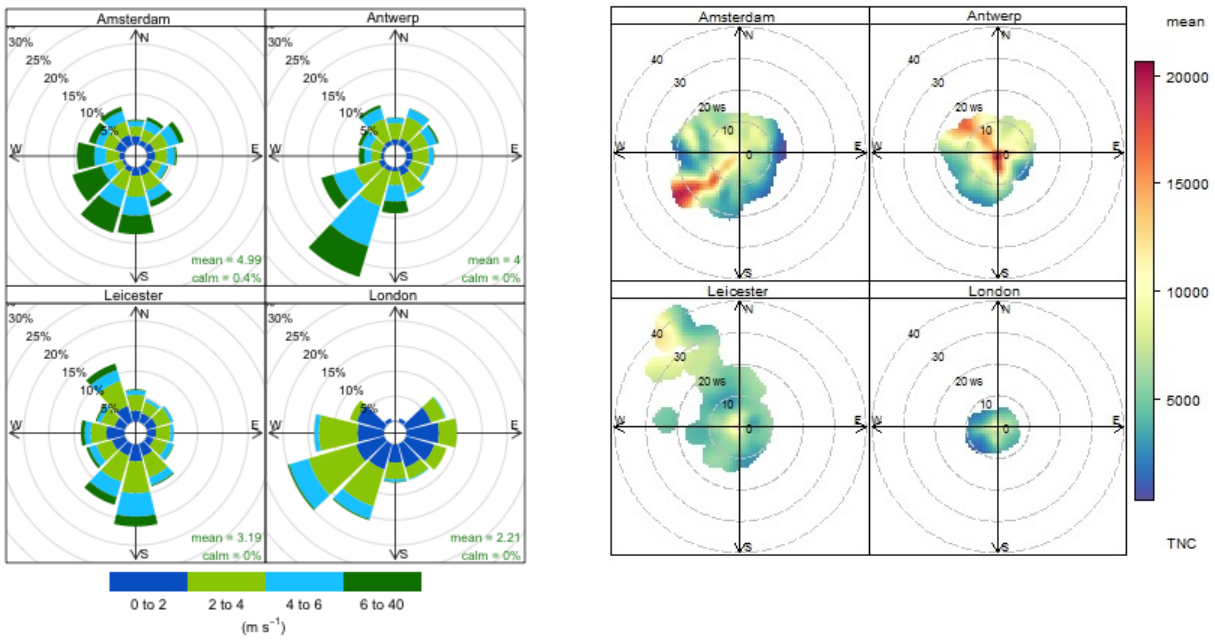


Figure 10: Wind roses (left) and polar plots of the average total number concentration (# $cm^{-3,}$ right) with respect to the experienced wind direction and speed for the considered monitoring periods in Amsterdam, Antwerp, Leicester and London.

When polar plots of the wind direction and speed averaged TNC are plotted per monitoring site (*Figure 10*, right panel), clear site-dependent effects are observed. While Leicester and London show relatively homogeneous TNC concentrations, independent of wind direction and wind speed, Amsterdam and Antwerp show significant TNC variation depending on the experienced wind fields. Moreover, based on the polar plots, the location of contributing UFP sources can be derived.

The Antwerp monitoring site is clearly positioned near a southern-located UFP source, namely the traffic-intensive *Plantin en Moretuslei*. This particular site experiences highest UFP concentations when wind speeds are low. At higher wind speeds, UFP emitted by the local traffic will be diluted,

resulting in lower UFP concentrations. An additional UFP input can be observed when the wind is blowing from the NW, where streets at the other side of the monitoring site are located, as was also observed in (Frijns et al., 2013b). Looking at the individual size classes, the source effect of the *Plantin en Moretuslei* is most apparent for the 20-30 and 30-50 nm size classes (results not shown). For the Amsterdam site, an average 38% increase in TNC can be observed under strong SW winds. Looking at the individual size classes, the increase in TNC for SW winds is only observed for the 10-20 and 20-30 nm size classes (not shown). This might be attributable to Schiphol airport emissions, in line with Keuken et al. (2015), who recently reported on a marked UFP number concentration increase in Amsterdam dominated by 10-20 nm sized particles during periods when the wind was blowing from Schiphol. The TNC increased by a factor of three at a monitoring station (Adamse Bos) located 7 km from Schiphol (Keuken et al., 2015). Another study reported on a comparable 4- to 5-fold increase in particle number concentrations at 8-10 km downwind of Los Angeles International airport (Hudda et al., 2014).

Taking into account the location of the Amsterdam site (AD1) at approximately 8 km downwind of Schiphol Airport (Figure 11), a non-traffic-related temporal variation of the 10-20 nm size range which persists throughout the weekends (see 3.3), and no clear relation between TNC and traffic-related pollutants (see 3.2), Schiphol seems to contribute to the urban UFP concentrations in Amsterdam. The TNC, measured at the AD1 monitoring site, was observed to increase by 34% when the wind was blowing from Schiphol (205-245°) compared to all other wind directions. As the city centre of Amsterdam is located downwind of Schiphol airport and south-westernly wind directions were experienced for 16% of the total monitoring time (5436 half-hourly values on a total of 34830 half-hourly values were between 205-245°), a significant attribution of Schiphol on citizens exposure in Amsterdam can be expected. Taking into account the 34% TNC increase and 16% occurrence of 205-245° wind directions, Schiphol airport determined 5.44% of TNC at the Amsterdam monitoring station near Vondelpark (city center of Amsterdam). Plotting the particle number concentrations of the smallest size class (10-20 nm) as a function of wind direction, this directional effect becomes much stronger as the 10-20 nm PNC is almost doubled (99% increase) when wind is blowing from 205-245° (*Figure 11*). Although less clear due to the much shorter monitoring period (2 weeks) and the possible upwind influence of Amsterdam itself, higher 10-20 nm concentrations are obtained as well at the trailer location (AD2T) when the wind was blowing from the SW. Taking into account the 16% occurrence of 205-245° wind directions, Schiphol airport accounted for 16% of the total experienced 10-20 nm particles at the AD1 monitoring site.

Figure 11: Locations of the fixed (AD1) and mobile unit (AD2T) monitoring sites at respectively 8 and 14 km from Schiphol airport, with pollution roses of the wind direction averaged (red) 10-20 nm concentration at the considered monitoring locations.

For Leicester, a slight increase in total particle number concentration can be observed for periods in which wind was blowing from the west (NW-SW). Potential contributors might be East Midland airport and Radcliffe Soar power station, which are both located at about 27 km NW of the considered monitoring site. The more distant source locations are reflected in the observed contribution at the monitoring site under high (>20 m s^{-1}) wind speeds. A north-south oriented main road (Welford Road) surrounded by residential areas is situated west of the Leicester monitoring station and a green area and Leicester University are situated east of the station. As the temporal variation shows a traffic-related diurnal variation, it can be assumed that the main road is contributing significantly to the measured particle number concentrations. The highest contribution in particle number concentration during western wind conditions was observed for the 20-30 nm size class (not shown).

London shows rather homogeneous particle number concentrations independent of the experienced wind fields. No clear effect of London Heathrow airport (± 35 km in western direction) or London city airport (± 8 km north) can be observed on the measured UFP concentrations. Only during strong and eastern wind conditions, can an increase in TNC be observed. This might be due to the Port of London, which is located at about 15 km in the eastern direction of the LO1 monitoring site. Previous studies already reported significant UFP contributions from shipping in coastal regions (González et al., 2011; Healy et al., 2009; Querol, 2011).

4. Conclusion

This study reports on a long time series (1-2 years) of total and size-resolved UFP number concentrations in four European urban background locations (Amsterdam, Antwerp, Leicester and London), supplemented with additional short-term mobile monitoring unit measurements (2-4

weeks), at each fixed site and an additional urban background location in Amsterdam, Antwerp and Leicester. With regard to urban UFP monitoring, this long-term time series provides important insights into the spatiotemporal variation of total and size-resolved ultrafine particles in urban environments. The obtained data coverage of the UFP measurements was similar to the coverage of more commonly measured pollutants (PM, NO_x, etc). From a practical point of view, including UFP measurements in current telemetric monitoring networks seems, therefore, feasible. Moreover, the co-located mobile monitoring unit provided a valuable addition to the fixed sites for harmonisation and validation purposes.

The fixed monitoring sites show comparable UFP size distributions with similar proportional contributions between the individual particle size classes (100-200 < 70-100 < 50-70 < 20-30 < 30-50 < 10-20 nm). Nevertheless, the size-resolved measurements enabled us to identify different contributing emission sources at different spatial scales. When comparing UFP size distributions between the various sites, better association was obtained between the larger UFP size classes (>50 nm). Larger particles, therefore, seem to be more uniform in space, which confirms the regional nature of these aerosols. Ambient UFP concentrations, in line with BC and NO_2, showed clear traffic-related diurnal variation with distinct morning and evening rush hour peaks on week days, but only a clear evening peak during the weekends. Apart from the diurnal traffic-related variation, new particle formation events were observed in the early afternoon, confirming the prevalence of nucleation events in northwestern Europe. Compared to the other sites, Antwerp experienced significantly higher total number concentrations owing to its proximity to the *Plantin en Moretuslei*, a busy access road into Antwerp, confirming road traffic as an important UFP source in urban environments.

In Amsterdam, a high and constant 10-20 nm contribution which persisted during the weekends (1), a significant additional input of mainly 10-20 nm sized UFPs under southwestly wind fields (2) and the lacking relationships between measured UFP concentrations and concentrations of typical traffic-related pollutants (2) confirmed Schiphol airport as a source of ultrafine particles, contributing to atmospheric UFP concentrations in the city centre of Amsterdam. Taking into account the frequency of southwestly wind fields, and the proportional increase of total and 10-20 nm sized particles, Schiphol airport was estimated to contribute to 5% of TNC and 16% of 10-20 nm particles measured at the Amsterdam site.

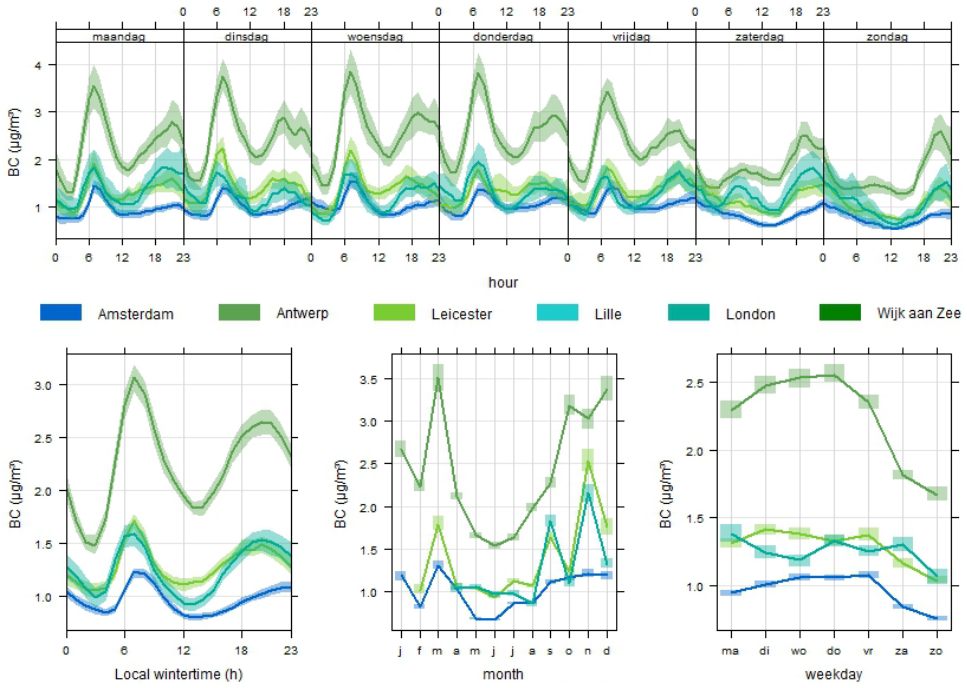
The spatial variation of UFPs inside teh respective cities was evaluated using simultaneous mobile monitoring unit measurements at additional urban background locations. Although covarying UFP concentrations were observed (r_s = 0.59 to 0.85), the absolute difference in terms of particle numbers have been shown to be significant (up to 38% and 49% for total- and size-resolved particle numbers, respectively). As all monitoring sites are classified as “urban background” locations, the observed differences will likely even increase at contrasting monitoring locations. This implies that the location of the UFP monitoring site is of primordial importance when evaluating citizen’s exposure to UFPs in urban environments. Compared to the total number concentration, size-resolved measurements have been shown to offer far more information on the type, origin and transformation processes of atmospheric aerosols. Moreover, by combining both total and size-resolved UFP instruments, instrument anomalies can be easily detected.

Acknowledgments

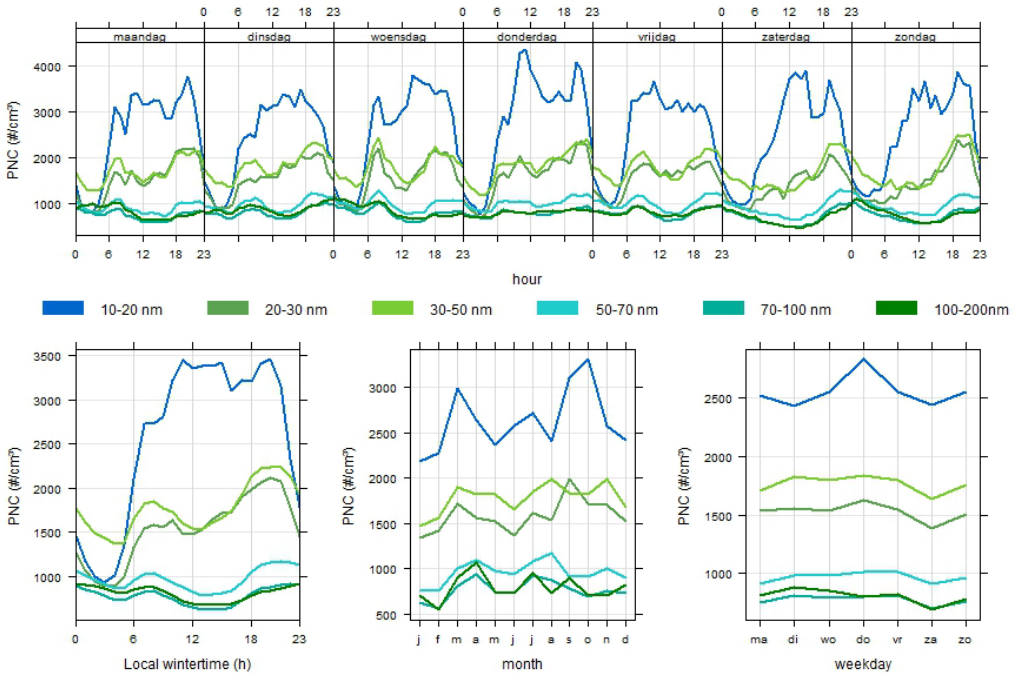
This research was funded by the Joint Air Quality Initiative (JOAQUIN) project, part of the EU Interreg IV-B NWE Program. The authors would like to thank the Health Service Amsterdam (GGD), Energy research Center of the Netherlands (ECN), DEFRA, the London Air Quality Network, ERG Kings College London and the Flanders Environment Agency (VMM) for their data and feedback.

Supplementary Material

Appendix 1: Temporal variation of BC (µg m^{-3}) for the considered monitoring stations (AD1, AP1, LE1 and LO1) at three different time scales (monthly, daily and hourly averages). The coloured zone represents the 95% confidence interval.



Appendix 2: Temporal variation of the particle number concentration (# cm^{-3}) obtained at the Amsterdam monitoring site (AD1) within the 10-20 nm, 20-30 nm, 30-50 nm, 50-70 nm, 70-100 nm and 100-200 nm size classes at three different time scales (monthly, daily and hourly averages).



Appendix 3: Coefficients of determination (COD, left) and Spearman Rank correlations (r_s, right) of the half-hourly size-resolved particle number concentrations between the respective monitoring sites. Only for Antwerp and Amsterdam, 10-20 nm size class measurements were available (SMPS).

COD 10-20 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.36	NA	NA
Amsterdam	0.36	0.00	NA	NA
Leicester	NA	NA	NA	NA
London	NA	NA	NA	NA

Spearman rank (r_s) 10-20 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	1.00	0.37	NA	NA
Amsterdam	0.37	1.00	NA	NA
Leicester	NA	NA	NA	NA
London	NA	NA	NA	NA

COD 20-30 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.33	0.35	0.44
Amsterdam	0.33	0.00	0.36	0.42
Leicester	0.35	0.36	0.00	0.40
London	0.44	0.42	0.40	0.00

Spearman rank (r_s) 20-30 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	1.00	0.36	0.31	0.11
Amsterdam	0.36	1.00	0.29	0.17
Leicester	0.31	0.29	1.00	0.34
London	0.11	0.17	0.34	1.00

COD 30-50 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.31	0.35	0.37
Amsterdam	0.31	0.00	0.35	0.35
Leicester	0.35	0.35	0.00	0.32
London	0.37	0.35	0.32	0.00

Spearman rank (r_s) 30-50 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	1.00	0.38	0.35	0.17
Amsterdam	0.38	1.00	0.25	0.15
Leicester	0.35	0.25	1.00	0.35
London	0.17	0.15	0.35	1.00

COD 50-70 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.30	0.34	0.39
Amsterdam	0.30	0.00	0.38	0.36
Leicester	0.34	0.38	0.00	0.35
London	0.39	0.36	0.35	0.00

Spearman rank (r_s) 50-70 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	1.00	0.48	0.39	0.21
Amsterdam	0.48	1.00	0.27	0.18
Leicester	0.39	0.27	1.00	0.38
London	0.21	0.18	0.38	1.00

COD 70-100 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.32	0.35	0.38
Amsterdam	0.32	0.00	0.41	0.37
Leicester	0.35	0.41	0.00	0.35
London	0.38	0.37	0.35	0.00

Spearman Rank (r_s) 70-100 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	1.00	0.60	0.39	0.17
Amsterdam	0.60	1.00	0.31	0.18
Leicester	0.39	0.31	1.00	0.36
London	0.17	0.18	0.36	1.00

COD 100-200 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	0.00	0.32	0.36	0.44
Amsterdam	0.32	0.00	0.38	0.40
Leicester	0.36	0.38	0.00	0.36
London	0.44	0.40	0.36	0.00

Spearman rank (r_s) 100-200 nm

	Antwerp	Amsterdam	Leicester	London
Antwerp	1.00	0.66	0.42	0.27
Amsterdam	0.66	1.00	0.38	0.28
Leicester	0.42	0.38	1.00	0.48
London	0.27	0.28	0.48	1.00

Bibliography

Brines, M., Dall'Osto, M., Beddows, D.C.S., Harrison, R.M., Gómez-Moreno, F., Núñez, L., Artíñano, B., Costabile, F., Gobbi, G.P., Salimi, F., Morawska, L., Sioutas, C., Querol, X., 2015. Traffic and nucleation events as main sources of ultrafine particles in high-insolation developed world cities. Atmospheric Chemistry and Physics 15, 5929–5945. doi:10.5194/acp-15-5929-2015

Carslaw, D.C., Ropkins, K., 2012. openair — An R package for air quality data analysis. Environmental Modelling & Software 27-28, 52–61. doi:10.1016/j.envsoft.2011.09.008

Carslaw, D.C., Ropkins, K., 2015. openair: Open-source tools for the analysis of air pollution data. Natural Environment Research Council.

Dall'Osto, M., Querol, X., Alastuey, A., O'Dowd, C., Harrison, R.M., Wenger, J., Gómez-Moreno, F.J., 2013. On the spatial distribution and evolution of ultrafine particles in Barcelona. Atmospheric Chemistry and Physics 13, 741–759. doi:10.5194/acp-13-741-2013

Donaldson, K., Stone, V., Clouter, A., Renwick, L., MacNee, W., 2001. Ultrafine particles. Occup Environ Med 58, 211–6, 199.

Frijns, E., Van Jo, L., Aerts, W., Brabers, R., Berghmans, P., 2013a. UFP instrument comparison at an urban background location in Antwerp.

Frijns, E., Van Laer, J., Berghmans, P., 2013b. Short-term intra-urban variability of UFP number concentration and size distribution.

Goel, A., Kumar, P., 2015. Characterisation of nanoparticle emissions and exposure at traffic intersections through fast–response mobile and sequential measurements. Atmos Environ 107, 374–390. doi:10.1016/j.atmosenv.2015.02.002

González, Y., Rodríguez, S., Guerra García, J.C., Trujillo, J.L., García, R., 2011. Ultrafine particles pollution in urban coastal air due to ship emissions. Atmos Environ 45, 4907–4914. doi:10.1016/j.atmosenv.2011.06.002

Harrison, R.M., Shi, J.P., Xi, S., Khan, A., Mark, D., Kinnersley, R., Yin, J., 2000. Measurement of number, mass and size distribution of particles in the atmosphere. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences 358, 2567–2580. doi:10.1098/rsta.2000.0669

Healy, R.M., O'Connor, I.P., Hellebust, S., Allanic, A., Sodeau, J.R., Wenger, J.C., 2009. Characterisation of single particles from in-port ship emissions. Atmos Environ 43, 6408–6414. doi:10.1016/j.atmosenv.2009.07.039

Hudda, N., Gould, T., Hartin, K., Larson, T.V., Fruin, S.A., 2014. Emissions from an international airport increase particle number concentrations 4-fold at 10 km downwind. Environ Sci Technol 48, 6628–6635. doi:10.1021/es5001566

Joaquin, 2015. Monitoring of ultrafine particles and black carbon. Joint Air Quality Initiative, Work Package 1 Action 1 and 3.

Kelly, F.J., Fussell, J.C., 2012. Size, source and chemical composition as determinants of toxicity attributable to ambient particulate matter. Atmos Environ 60, 504–526. doi:10.1016/j.atmosenv.2012.06.039

Keuken, M.P., Moerman, M., Zandveld, P., Henzing, J.S., Hoek, G., 2015. Total and size-resolved particle number and black carbon concentrations in urban areas near Schiphol airport (the Netherlands). Atmos Environ 104, 132–142. doi:10.1016/j.atmosenv.2015.01.015

Kozawa, K.H., Winer, A.M., Fruin, S.A., 2012. Ultrafine particle size distributions near freeways: Effects of differing wind directions on exposure. Atmos Environ 63, 250–260. doi:10.1016/j.atmosenv.2012.09.045

Krudysz, M., Moore, K., Geller, M., Sioutas, C., Froines, J., 2009. Intra-community spatial variability of particulate matter size distributions in Southern California/Los Angeles. Atmospheric Chemistry and Physics 9, 1061–1075. doi:10.5194/acp-9-1061-2009

Kulmala, M., Kerminen, V., 2008. On the formation and growth of atmospheric nanoparticles. Atmos Res 90, 132–150. doi:10.1016/j.atmosres.2008.01.005

Kumar, P., Morawska, L., Birmili, W., Paasonen, P., Hu, M., Kulmala, M., Harrison, R.M., Norford, L., Britter, R., 2014. Ultrafine particles in cities. Environ Int 66, 1–10. doi:10.1016/j.envint.2014.01.013

Mishra, V.K., Kumar, P., Van Poppel, M., Bleux, N., Frijns, E., Reggente, M., Berghmans, P., Int Panis, L., Samson, R., 2012. Wintertime spatio-temporal variation of ultrafine particles in a Belgian city. Sci Total Environ 431, 307–313. doi:10.1016/j.scitotenv.2012.05.054

Panteliadis, P., Strak, M., Hoek, G., Weijers, E., van der Zee, S., Dijkema, M., 2014. Implementation of a low emission zone and evaluation of effects on air quality by long-term monitoring. Atmos Environ 86, 113–119. doi:10.1016/j.atmosenv.2013.12.035

Pey, J., Rodríguez, S., Querol, X., Alastuey, A., Moreno, T., Putaud, J.P., Van Dingenen, R., 2008. Variations of urban aerosols in the western Mediterranean. Atmos Environ 42, 9052–9062. doi:10.1016/j.atmosenv.2008.09.049

Querol, X., 2011. Ultrafines in the atmosphere.

R Development Core Team, 2015. R: A language and environment for statistical computing. R Foundation for statistical computing.

Viana, M., Rivas, I., Reche, C., Fonseca, A.S., Pérez, N., Querol, X., Alastuey, A., Álvarez-Pedrerol, M., Sunyer, J., 2015. Field comparison of portable and stationary instruments for outdoor urban air exposure assessments. Atmos Environ. doi:10.1016/j.atmosenv.2015.10.076

VMM, 2014. Intra-urban variability of ultrafine particles in Antwerp (February and October 2013).

Von Bismarck-Osten, C., Birmili, W., Ketzel, M., Massling, A., Petäjä, T., Weber, S., 2013. Characterization of parameters influencing the spatio-temporal variability of urban particle number size distributions in four European cities. Atmos Environ 77, 415–429. doi:10.1016/j.atmosenv.2013.05.029

Vu, T.V., Delgado-Saborit, J.M., Harrison, R.M., 2015. Review: Particle number size distributions from seven major sources and implications for source apportionment studies. Atmos Environ 122, 114–132. doi:10.1016/j.atmosenv.2015.09.027

Wang, D., Guo, H., Cheung, K., Gan, F., 2014. Observation of nucleation mode particle burst and new particle formation events at an urban site in Hong Kong. Atmos Environ 99, 196–205. doi:10.1016/j.atmosenv.2014.09.074

Zhu, Y., Hinds, W.C., Kim, S., Shen, S., Sioutas, C., 2002. Study of ultrafine particles near a major highway with heavy-duty diesel traffic. Atmos Environ 36, 4323–4335. doi:10.1016/S1352-2310(02)00354-0