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Monitoring PM_{10} and Ultrafine Particles in Urban Environments Using Mobile Measurements

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ABSTRACT

Mobile platforms are increasingly used to acquire air quality data at a high spatial and temporal resolution in complex urban environments. As such, mobile measurements provide a solution for short-term studies to acquire a spatially spread data set that would not be feasible if using stationary measurements. Mobile monitoring campaigns were carried out with a bicycle platform at two different urban locations, consisting of 20 and 24 repeated runs along a fixed route over a three-week period. The measurement runs were carried out on different days and at different times of the day, without systematical temporal coverage. Significant differences in UFP concentration were found within the day and between days, and also between several streets along the measurement route. These differences were related to traffic intensity and street characteristics. In contrast, PM_{10} concentrations differed between measurement days, but the within-day variability of PM_{10} was mostly non-significant. Additionally, the spatial variability was limited and the PM_{10} concentrations were only significantly different between busy streets, with high concentrations, and quiet background streets, with low ones. The results indicate that for most streets the number of runs was sufficient to give a good approximation of median daytime UFP concentration levels for the measurement period, and for some streets this number could even be reduced to less than 10. However, for PM_{10} a higher number of runs is needed, and this may be attributed to the significant background contribution to the roadside PM_{10} concentration, and the high variability of this. We conclude that a limited set of mobile measurements makes it possible to map locations with systematically higher or lower UFP and PM_{10} concentrations in urban environments.

Keywords: Particulate matter; UFP; Air quality; Mapping; Mobile monitoring; Pollution.

INTRODUCTION

Particulate air pollution is a mixture of particles that vary in number, size, shape, surface area, chemical composition, solubility and origin, where the size distribution is typically trimodal, including coarse particles (aerodynamic diameter > 2.5 μm), fine particles (aerodynamic diameter between 0.1 and 2.5 μm) and ultra-fine particles (UFP, aerodynamic diameter < 0.1 μm) (Pope and Dockery, 2006). This study focuses on the ultra-fine fraction and the coarse fraction with an aerodynamic diameter of < 10 μm (PM_{10}). PM_{10} is primarily derived from suspension and resuspension of solid material, and contributes greatly to the mass of the total suspended particles in urban environments. In contrast, UFP contribute little to the mass of the total suspended particles but they are highly abundant. The urban particulate cloud is constantly receiving UFP from primary emissions from combustion sources in transportation, industries and power generation, and by secondary formation by atmospheric photochemical reactions and conversion processes (Westerdahl *et al.*, 2005; Seinfeld and Pandis, 2006). UFPs have a transient nature with short life times (minutes to hours) and rapidly grow through atmospheric processes of coagulation and/or condensation to larger complex aggregates (Pope and Dockery, 2006). Therefore, the highest concentrations of ultrafine particles are found in the vicinity of the primary sources, for example, near busy roads where particle number concentrations are typically between 10^4 and 10^6 particles/cm^3 depending on driving speed, fleet composition and meteorology (Nikolova *et al.*, 2011) and ultrafine particle number concentrations decrease rapidly with distance from the emission sources (Zhu *et al.*, 2002; Hagler *et al.*, 2010). Therefore important differences, in space and time, of UFP concentrations between urban micro-environments are induced (More *et al.*, 2009; Hudda *et al.*, 2010).

Small-scale variations in PM_{10} concentrations have also been reported elaborately in the literature. Temporal patterns of PM_{10} have been observed over seasons (Monn *et al.*, 1997; Liu *et al.*, 2003), weeks (Monkkonen *et al.*, 2004), days (Roosli *et al.*, 2001) and hours of the day (Gomiscek

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et al., 2004) in urban settings and were attributed to the temporal dynamics of atmospheric conditions and the PM_{10} sources. The spatial variability of PM_{10} within the urban environment differs in the literature from rather limited (Roosli *et al.*, 2000) to substantial (Chan *et al.*, 2001; Wilson *et al.*, 2006), which may be predominantly caused by the spatial heterogeneity of the main (traffic) sources.

The epidemiological importance of particles and their high concentrations in urban environments have resulted in permanent or temporary surveillance of particle concentrations in complex urban terrain. Mobile measurements are frequently applied as an complementary tool to the stationary air quality measurements at fixed locations, because fixed stations are not capable to depict the full spatial distribution of air pollution over the extent of an urban area (Kaur *et al.*, 2007). For example, Berghmans *et al.* (2009) estimated the exposure of a cyclist to (ultrafine) particles in an urban environment by monitoring particle concentrations with an instrumented bicycle. Westerdahl *et al.* (2005) integrated air pollution measurement instrumentation with a mobile platform to characterize UFP and other pollutants (NO_x, Black Carbon) in the Los Angeles area. Kaur *et al.* (2005) quantified the personal exposure of street canyon intersection users to UFP, $PM_{2.5}$ and CO in Central London by mobile data collection by volunteers who travelled a predefined route via five modes of transport at different times of the day, and Zhu *et al.* (2008) developed and tested a mobile laboratory to facilitate concurrent measurements of near real time, in-vehicle concentration and size distribution of UFP, other traffic-related air pollutants for health effect studies. Mobile measurements are also applied for high resolution mapping of the spatial variability of air pollution (Hagler *et al.*, 2010) and for the characterization of the local source contributions to the particulate air pollution in urban environments. Mobile monitoring of traffic related pollutants have been used to develop models (e.g., land use regression (LUR) models) that allow a further investigation of spatial pollutant patterns (Buonocore *et al.*, 2009; Larson *et al.*, 2009; Zwack *et al.*, 2011). Besides these controlled set-ups, mobile monitoring can also be the core of monitoring schemes in which data are collected by volunteers during their normal daily routines.

In view of the increasing application of mobile measurements for urban air quality monitoring, and the potential for monitoring schemes by volunteers to cover large areas of a city, we carried out two mobile monitoring campaigns to investigate to what extent a limited set of mobile measurements spread over different days and different times of the day allows to draw conclusions on spatio-temporal variation in urban air pollution and to map urban air quality.

MATERIAL AND METHODS

Study Area and Monitoring Protocol

The mobile measurements were performed at two locations, Antwerp (51°12'N, 4°26'E) and Mol (51°11'N, 5°07'E), Belgium. Antwerp is a medium-sized city (480000 inhabitants, 985 inhabitants/km^2), Mol is a provincial town (34000 inhabitants, 299 inhabitants/km^2). At both locations, a fixed route was defined using cycling paths or the right side of the road. The route in Antwerp was approximately 5 km long, the route in Mol 10 km (Figs. 1(a)–1(b)). Average travelling time for the entire route were approximately 25 minutes for Antwerp and 50 minutes for Mol. Although the major part of the mobile routes were located in residential area, streets of differing configuration and with differing traffic dynamics were included in this study. The results presented in this study are focussed upon a selection of six streets in Antwerp and Mol which reflect the variation in traffic density, driving speed and street configuration (Table 1). A recreational area (Kleinendijk) with very low traffic density was included along the route in Mol. After a data quality control, 24 runs on 8 days in the period between March 16 and April 8, 2009 were withheld in Antwerp (Table 2). In Mol a total of 20 runs were performed on 10 measurement days between April 7 and April 23, 2010 (Table 2). The monitoring hours ranged from 6 a.m. until 6 p.m., but most of the runs were made between 10 a.m. and 4 p.m. The repetition frequency of the measurement runs (number of runs per day, time of the day) was partially determined by practical and organizational circumstances. This led to a non-uniform distribution of the measurements over the day (Fig. 1(c)). All measurements were carried out during working days.

Mobile Platform

Portable UFP and PM monitors were installed on a bicycle which was additionally equipped with a GPS (Garmin Forerunner) to register measurement location and a smartphone (Openmoko FreeRunner) to synchronize the sensors and to communicate the data to the central database. A TSI P-Trak ultrafine particle counter (model number 8525) was used to measure the number concentration of ultrafine particles within a range from 0 to $5 \cdot 10^5$ particles/cm^3 at a temporal resolution of 1 sec. The P-Trak is based on the condensation particle counting technique using isopropyl alcohol (TSI, http://www.tsi.com). The particle size range is from 0.02 to 1 μm, and thus beyond the upper limit of ultrafine particles of 0.1 μm. The flow rate of the P-Trak was 0.7 L/min, the temperature during operation was well within the limits provided by the manufacturer, and alcohol saturation of the wicks was guaranteed. Laboratory tests of P-Trak to a TSI model 3022 CPC showed an excellent agreement (99% R^2), but the P-Trak concentrations were about 30% lower due to its lower detection limit of 0.02 μm which is higher than the 0.007 μm detection limit of CPC (Chan *et al.*, 2004; Wallace *et al.*, 2011, and reference therein). In the first part of this study, P-Trak measurements are not compared to absolute UFP number concentrations, so this bias would not affect the conclusions. In the next part, absolute UFP number concentrations are compared between Antwerp and Mol, and with UFP concentrations reported in the literature. In the latter comparison, instrument specific bias may occur. Information on the instrumentation is carefully added to avoid erroneous interpretation due to differences in instrumentation.

A DustTrak DRX 8534 was used in Antwerp for

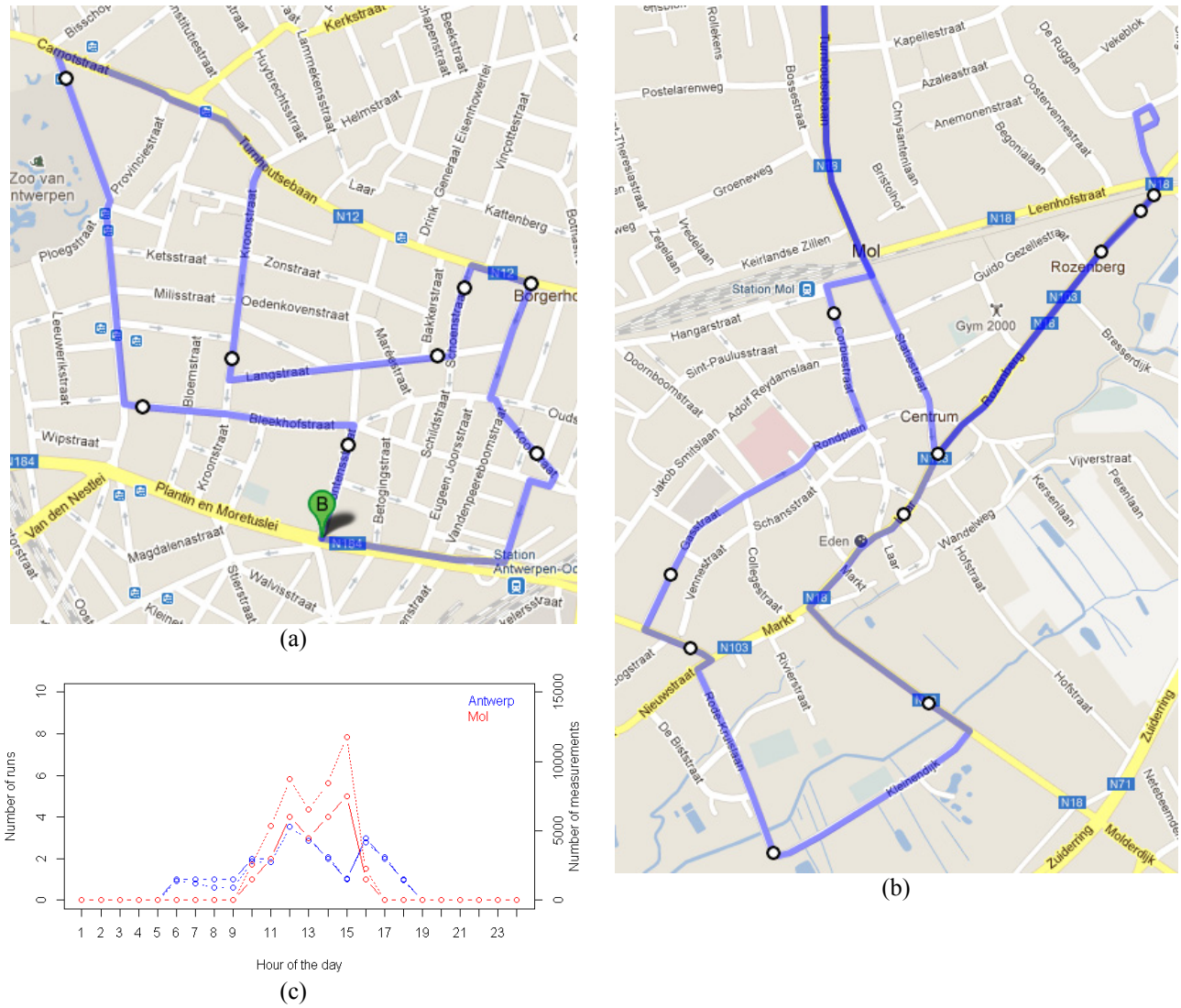

Fig. 1. The mobile monitoring routes in Antwerp (a) and Mol (b), and an overview of the number of runs (full line) and the number of measurement data (dotted line) in function of time of the day (c).

Table 1. Overview of some street characteristics at a selection of streets along the mobile monitoring routes in Antwerp and Mol.

	Street name	Speed limit	Configuration	Traffic density [day^{-1}]*		
				light	heavy	total
Antwerp	Bleekhofstraat	50 km/h, 30 km/h	1 lane	4 074	8	4 082
	Provinciestraat	50 km/h	1 lane, street canyon	13 495	4	13 499
	Carnotstraat	50 km/h	1 lane, street canyon, separate biking lane	21 396	119	22 515
	Plantin en Moretuslei	70 km/h	2 lanes, separate biking lane	42 961	420	43 381
	Langstraat	50 km/h	1 lane	no data	no data	no data
	Kroonstraat	50 km/h	1 lane	3 046	0	3 046
Mol	Turnhoutsebaan	70 km/h, 50 km/h	1 lane, separate biking lane, commercial and residential	9 419	297	9716
	Statiestraat	50 km/h	1 lane, street canyon, residential and commercial	4 143	262	4 405
	Rozenberg	50 km/h	1 lane, separate biking lane, residential	5 603	115	5 718
	Kleinendijk	50 km/h	1 lane, green (recreational) zone	no data	no data	no data
	Gasstraat	50 km/h	1 lane, residential	1 852	0	1 852
	Voogdijstraat	50 km/h	1 lane, street canyon	6 023	326	6 349

* data source: Traffic Centre Flanders.

Table 2. Overview of the measurement runs.

Antwerp			Mol		
Date	Run	Starting Hour	Date	Run	Starting hour
2009/03/16	1	12:15:54	2010/04/07	1	14:21
	2	12:44:44		2	15:27
	3	13:09:51	2010/04/08	3	14:05
2009/03/27	4	16:08:22		4	14:54
	5	16:44:12	2010/04/12	5	15:04
	6	16:58:29		6	15:55
2009/03/30	7	11:57:50	2010/04/14	7	12:16
2009/03/31	8	10:38:40		8	13:02
	9	11:39:53	2010/04/15	9	12:32
2009/04/02	10	12:04:04		10	13:18
	11	13:21:19	2010/04/16	11	12:46
	12	14:17:25		12	13:38
2009/04/03	13	10:18:16	2010/04/19	13	14:18
	14	11:14:45		14	15:12
	15	13:13:03	2010/04/20	15	10:40
	16	14:09:28		16	11:33
2009/04/06	17	15:15:00	2010/04/22	17	15:23
	18	16:09:16		18	16:14
	19	17:04:34	2010/04/23	19	11:17
	20	18:00:54		20	12:06
2009/04/08	21	5:53:05			
	22	6:53:20			
	23	7:49:33			
	24	8:48:14			

measuring PM_{10} mass concentration ($\mu g/m^3$), whereas in Mol the PM_{10} concentration was measured by a GRIMM 1.108 Dust monitor. The instruments were recently calibrated and operated at flow rates of 1.7 L/min and 1.2 L/min, respectively. The outdoor temperature was within the operating temperature range. PM_{10} concentrations were measured at a 1 and 6 second resolution in Antwerp and Mol, respectively. In Antwerp, two DustTrak instruments were used at different occasions, and a correction factor derived from a linear regression ($R^2 = 0.99$) between both instruments during 5 days of simultaneous measurements was used. We note that DustTrak may significantly overestimate actual PM_{10} concentration (Chung *et al.*, 2001). Literature reports on an overestimation of real aerosol concentrations by a factor two to three by the DustTrak (e.g., Chung *et al.*, 2001; Braniš and Větvička, 2010). We did not introduce additional uncertainty by rescaling the data by an arbitrary coefficient because the measurement were primarily used a relative way. The effect of the relative humidity on the PM_{10} measurements was reduced by avoiding to conduct measurement in high humidity conditions. The relative humidity ranged between 45 and 88%, and was below the 95% critical threshold reported in the DustTrak user's manual. A similar mobile platform setup as the mobile setup for Mol was shown to have a robust performance in a similar urban environment (Berghmans *et al.*, 2009). Comparison of PM_{10} data from Antwerp and Mol was not feasible given the different instrumentation at both sites.

Analyses

An overall data inspection of the sets of mobile measurements from Antwerp and Mol was first made, without stratifying for time or street. A cumulative density function of the measurements at both study sites was calculated and pollutant concentrations were statistically compared using a non-parametric Kruskal-Wallis test at a critical value (α) of 0.05 (Kruskal and Wallis, 1952). This is a non-parametric alternative to a one-way ANOVA. The Kruskal-Wallis test investigates whether the sampled populations have the same median.

Furthermore, an analysis of the temporal and spatial variability of the air quality measurements was conducted to assess the potential of using mobile measurements to distinguish between episodes of low or high particle concentrations, and between locations (streets) of contrasting particle concentrations. A post-hoc multiple comparison between measurement days, time of the day, or street was performed using the results of a Kruskal-Wallis test with a Bonferroni adjustment of the critical value to compensate for multiple comparison. Given the relative nature of the spatio-temporal assessment, errors due to instrumentation differences (e.g., DustTrak DRX 8534 vs. GRIMM 1.108) and differences in meteorological conditions were reduced.

Given the spatio-temporal dynamics of the pollutants under investigation and the fact that mobile measurements provide snap-shots of pollutant concentrations in space and time, an experiment was conducted on the data to investigate the representativeness. The research question addressed was how many mobile runs are needed to obtain a representative estimation of the street-level air pollution in a confined area.

In this study, representative means that the estimation is reasonably similar (± 15%) to the value obtained from the entire mobile measurement campaign. The representativeness is thus constrained by the space and time of data collection, which was biased toward daytime during one month of the year in an urban environment, and not necessarily representative for other periods or locations. We mainly focused on the time of the day that (the majority of the) people are really exposed, and do not include night-time. Additionally, we assessed how sensitive the results are for the timing of those runs.

First, all the data were used to calculate an aggregated median pollutant concentration per street over all the measurement runs. We preferred to use the median concentration because air quality measurements are not normally distributed and skewed, so the median is a more representative central tendency measure than the mean. Subsequently, median pollutant concentrations were calculated per street based on mobile measurements of a cumulatively growing data volume that was obtained by a cumulative addition of measurement data from randomly selected runs (random selection without replacement). The median pollution concentrations in function of the increasing number of runs were compared to the overall median value to see after how many sampled runs the medians converged to the overall median. Convergence is obtained when the median of the sampled runs deviates less than 15% from the overall median, and does so consistently when adding new runs. The pseudo-code (Table 3) for this experiment is given below. The pseudo-code was run a high number of times (10000 iterations) to guarantee a high number of possible combinations of runs. The number of runs needed for convergence for each combination of randomly selected runs was plotted in a density plot. The overall median concentration may deviate substantially from an absolute concentration which would be measured by stationary reference devices.

A qualitative assessment on the temporal coverage of the runs in relation to the number of runs needed to allow for convergence was added. Here, two contrasting sequences of run combinations were highlighted: firstly a sequence by which the temporal coverage of the first selected runs was low, and secondly a sequence by which the temporal coverage of the first selected runs was high. Of course, when many runs are combined the temporal coverage of both sequences becomes more similar, to be identical when all the runs are combined. However, for combinations of a limited number of runs (i.e., i from pseudo-code is low) the temporal coverage may differ substantially. The two highlighted cases are referred to as the high similarity sequence and the low similarity sequence. For Antwerp, the first sequence (high similarity) was developed by adding runs made at similar times of the day with a higher likelihood, whereas the second sequence was obtained by adding runs with dissimilar times of the day with a higher likelihood. This analysis was not performed on the data from Mol because the limited coverage of the hours of the day.

RESULTS

Summary of the Mobile Measurements

Summary statistics of pollutant concentrations are given in Table 4, and their cumulative density functions are plotted in Fig. 2. The median UFP concentrations in Antwerp (15600 pt/cm^3) were significantly higher than UFP concentrations in Mol (median of 10500 pt/cm^3, Chi-sq = 8130, $p < 0.01$). The difference in UFP concentration between Antwerp and Mol was slightly higher at low percentiles than for high percentiles. The first quartile UFP concentration in Antwerp was 75% higher than the first quartile concentration of Mol (11200 pt/cm^3 versus 6370 pt/cm^3), whereas the difference between the third quartile concentrations was 40% (24600 pt/cm^3 versus 17500 pt/cm^3). The maximal concentration reached the upper detection limit of 500000 pt/cm^3 in Antwerp and was 379000 pt/cm^3 in Mol.

The median PM_{10} concentration in Antwerp was 83 μg/m^3 and the interquartile range of the measurements was 88 μg/m^3, suggesting a high variability between the measurements. In Mol, the median PM_{10} concentration was 34 μg/m^3 and the interquartile range reached 31 μg/m^3.

Spatio-temporal Analysis

A clear distinction is observed between the spatio-temporal dynamics of UFP and PM_{10}, respectively by plotting the median UFP and PM_{10} concentrations for a selection of streets (Fig. 3). UFP showed a high variability in space (between streets) and time (between measurement runs). The temporal changes of UFP concentration in Antwerp and Mol were present in both busy and quiet streets. High spatial differences between streets were also observed, with

Table 3. Pseudo-code of the data experiment.

Input: Mobile measurements made during k runs along the same route of UFP and PM_{10}
Output: Median pollutant concentration in function of the number of runs

1. randomly permute the k runs;
2. alculate overall median UFP and overall median PM_{10};
3. for i = 1 to k
   a. select pollutant data from the i first permuted runs;
   b.calculate the median UFP and median PM_{10} concentration, save;
   c. calculate the difference of median UFP and median PM_{10} with overall median UFP and overall median PM_{10}, respectively (as %), save;
   end
4. identify the number of runs at convergence; save

Table 4. Descriptive statistics of the UFP and PM_{10} concentrations measured along the routes in Antwerp and Mol.

Location	Period	Pollutant	Average	25%	Median	75%	IQR*
Antwerp	2009/03/16–2009/04/08	UFP	21722	11200	15600	24600	13400
		PM_{10}	97	37	83	125	88
Mol	2010/04/07–2010/04/23	UFP	16040	6370	10500	17500	11130
		PM_{10}	45	25	34	56	31

* Interquartile range

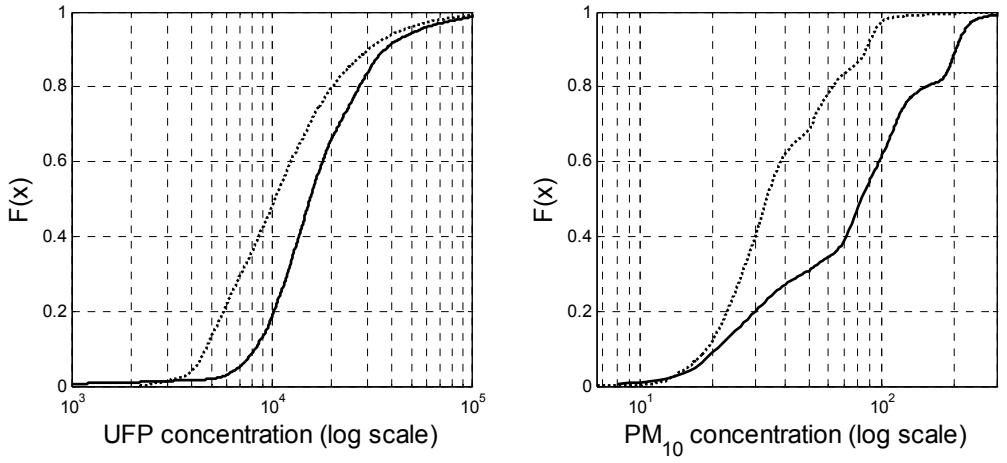

Fig. 2. Cumulative density functions of UFP and PM_{10} in Antwerp (solid line) and Mol (dotted line).

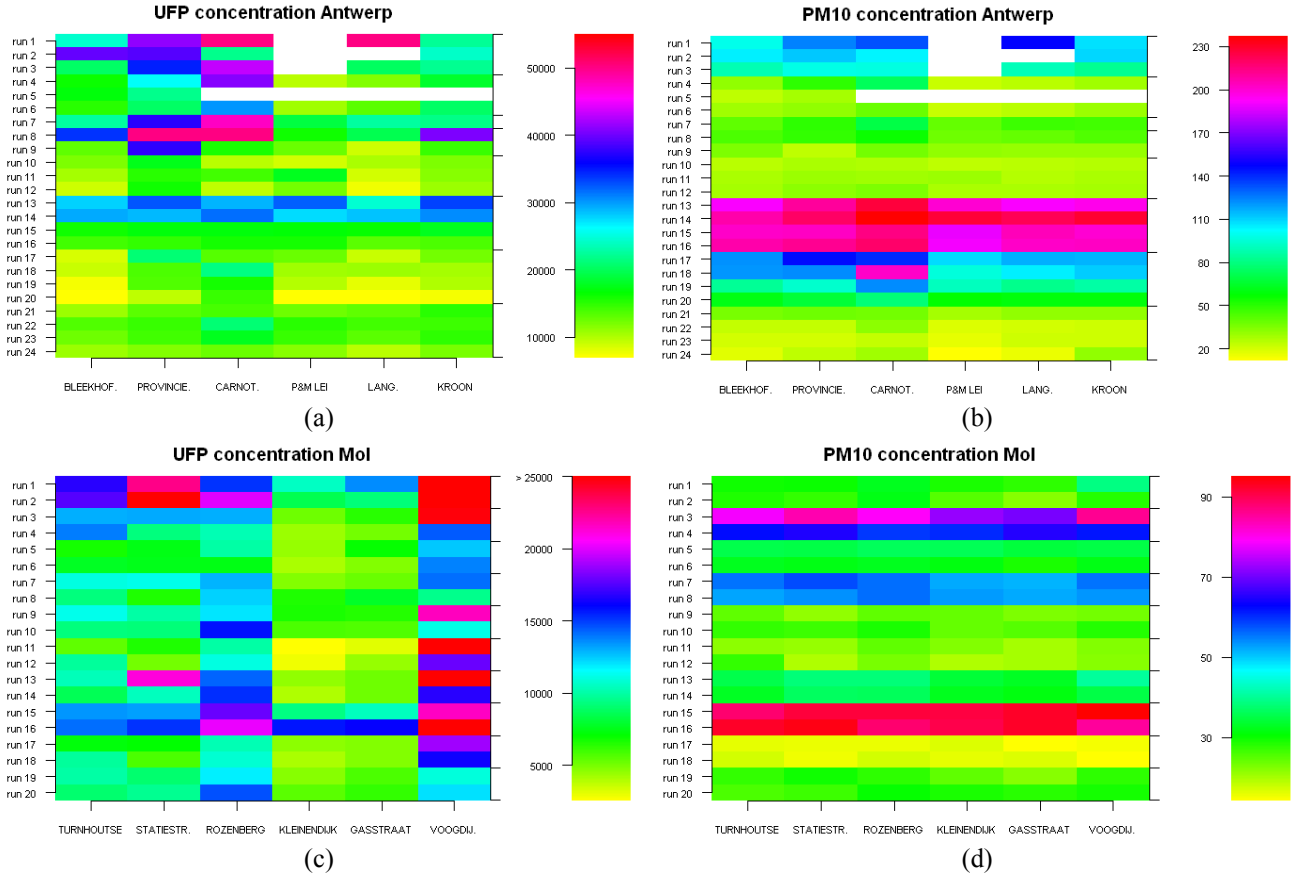

Fig. 3. Median UFP and PM_{10} concentrations for the different runs (y-axis) at a selection of streets (x-axis) in Antwerp and Mol. The different measurement days are indicated by the ticks on the second y-axis. The colours are scaled between the extremes and differ between the plots. White pixels represent no data.

high concentrations for Provinciestraat in Antwerp and for Voogdijstraat in Mol. Other streets consistently showed lower UFP concentrations. In contrast to the spatio-temporal pattern of UFP, PM_{10} showed a quite different pattern. The variability in space for PM_{10} was very limited compared to the variability in time. The PM_{10} concentrations did not differ a lot between streets. The variability in time was high between runs carried out on different measurement days, but much lower between runs carried out at different hours within the same measurement day.

A statistical analysis of the temporal pattern of UFP revealed both in Antwerp and in Mol significant differences in UFP concentration between the measurement runs (Chi-sq. = 16240, $p < 0.01$ for Antwerp, and Chi-sq. = 9386, $p < 0.01$ for Mol), but also between some of the mobile runs on the same day. In Antwerp, for example, UFP concentrations were significantly higher on March 16, 2009 than on April 6, 2009. On April 6, 2009 the UFP concentrations were significantly higher during the first run in contrast to the remaining three runs. UFP concentrations showed a high temporal variability, with significant changes in concentrations between hours of the same day.

The temporal variability of PM_{10} concentrations was high between measurement dates. Statistical tests indicated significant differences between several measurement dates (Chi-sq. = 31076, $p < 0.01$, Chi-sq. = 7159, $p < 0.01$, for Antwerp and Mol, respectively). The variability between PM_{10} concentrations of the mobile runs on a given day were, however, generally less pronounced than for UFP concentrations, and were often non-significant.

The average PM_{10} concentration of each run was compared with the hourly averaged PM_{10} measurements from stations of the Flemish Environmental Agency (VMM) located in the vicinity of the mobile routes (VMM station Borgerhout 42R801, located on the biking route in Antwerp, and VMM station Dessel 42N016, 5 km from the biking route in Mol). The PM_{10} concentrations measured at the reference stations followed a similar trend as the average PM_{10} concentrations from mobile measurements in Antwerp (Fig. 4(a), pearson correlation of 0.90), however, absolute values measured at the reference stations were generally (far) below the measured concentrations at the streets. The concentrations at the VMM station ranged between 8 and 139 $\mu g/m^3$. In periods of high PM_{10} concentration they were almost half of the concentration measured by the mobile platform. In Dessel near Mol, the concentrations from the VMM station were considerably lower, ranging between 14 and 58 $\mu g/m^3$, and also the trend of the measurements differed considerably between the VMM data and the mobile data (pearson correlation of 0.45, Fig. 4(b)). PM_{10} concentrations at the reference station were particularly low in comparison with the average concentration of run 3 and 4, which could possibly be attributed to a time lag between the strong decreasing trend in PM_{10} concentration at the VMM station and Mol city center on that day (2010/04/08, Fig. 4(c)). The decrease in PM_{10} concentration at the reference station was caused by precipitation (0.65 mm/hour between 6:00 and 15:00).

A statistical analysis of the spatial pattern of UFP and PM_{10} concentrations revealed significant concentration differences between the streets of the route in Antwerp (Fig. 5, Chi-sq. = 2711, $p < 0.01$). The highest UFP concentrations were measured in Carnotstraat and Provinciestraat, followed by Kroonstraat, which were significantly higher than the UFP concentrations in Plantin en Moretuslei and Bleekhofstraat. The lowest UFP concentrations were found in Langstraat. In Mol, significantly different UFP concentrations were also found between several streets (Fig. 5, Chi-sq. = 9766, $p < 0.01$). The UFP concentration in Voogdijstraat was significantly higher than in all the other streets along the route, whereas the concentrations at Kleinendijk were significantly lower than in the other streets. Between both extremes, Voogdijstraat and Kleinendijk, several other streets differed significantly in UFP concentration.

The spatial variability of PM_{10} was lower, but overall significant in Antwerp (Chi-sq. = 626, $p < 0.01$) and Mol (Chi-sq. = 46, $p < 0.01$). In Antwerp, measured PM_{10} concentrations were significantly higher in Carnotstraat in comparison to Provinciestraat and Kroonstraat, where the PM_{10} concentration was significantly higher than in all the other streets along the route (not shown). The PM_{10} concentration in these streets, however, were not significantly different. The differences in PM_{10} concentrations between streets in Mol were overall significant, but the multiple comparison test revealed that the this significance was solely attributed to the difference in the PM_{10} concentration at Gasstraat which was significantly lower than in Statiestraat and Rozenberg. The streetwise comparison did not reveal any other significant difference.

Experiment: How Many Runs are Needed?

One experiment was conducted, by which mobile runs were randomly and cumulatively added to calculate median street UFP and PM_{10} concentrations from a cumulatively growing dataset of mobile measurements. The data set is quite sparse, and the time intervals for the mobile runs were different on every day. In Antwerp, the total daytime interval in which mobile runs were carried out, was from 6 a.m. until 6 p.m. However, for many hours of the day only one or two runs were available (Fig. 1(c)). In Mol, measurements were performed on a daytime interval from 11 a.m. until 3 p.m.

Results indicated differences between UFP and PM_{10} at both study sites (Fig. 6). For the analysis of the UFP data set, the maximal number of runs required for UFP for convergence was 18 (out of 24) for Antwerp and 16 (out of 20) for Mol. At both measurement locations, however, 75% of the combinations convergence was already reached with a number of runs that was approximately 70% lower (7–8 instead of 24 runs in Antwerp, 6–7 instead of 20 runs in Mol). Inspection of the results for the individual streets showed differences between the streets. In Antwerp, an early peak around 2-3 runs was observed in the density curve for Plantin en Moretuslei and Kroonstraat, whereas this peak was shifted toward a higher number of runs (5 to 8) for Bleekhofstraat, Provinciestraat and Langstraat, and a much higher value (15) for Carnotstraat. For Mol, most streets showed a similar curve, except for Statiestraat and Voogdijstraat where the UFP measurements only converged after a higher number of repeated measurement runs.

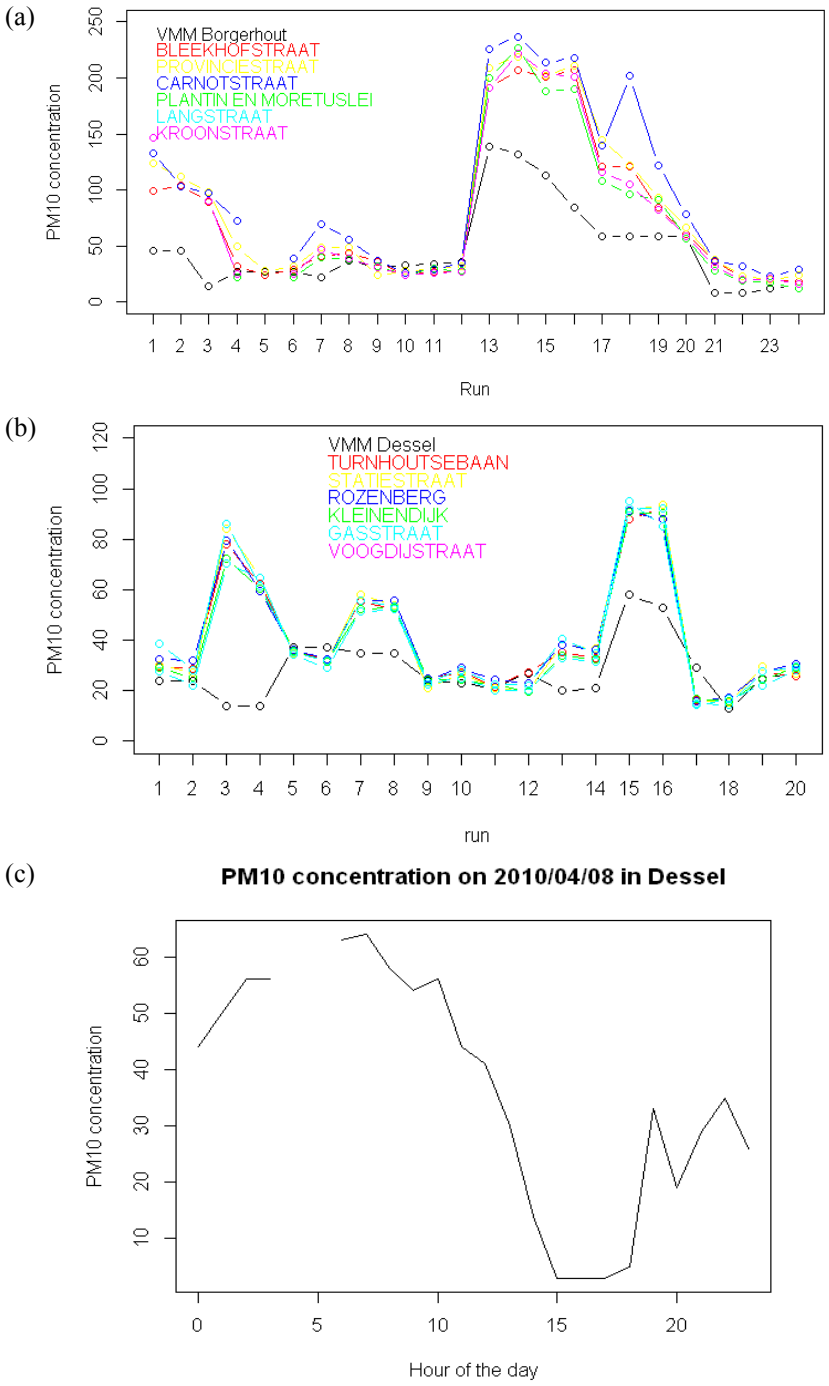

Fig. 4. Temporal fluctuations of PM_{10} concentrations from mobile measurements at a selection of streets in Antwerp (a) and in Mol (b) and at reference stations in the near vicinity of the routes (station Borgerhout and station Dessel, Flemish Environmental Agency). The difference between the mobile and stationary reference value during run 3 and 4 is caused by a steep decrease in PM_{10} when mobile measurements were conducted between 14:00 and 16:00 (c).

The differences between Antwerp and Mol were higher for PM_{10} than for UFP, yet a higher number of repeated measurement runs were required at both locations. The maximum number of runs needed for convergence reached values of 20 and 15 for Antwerp and Mol, respectively. 75% of the combinations of runs reached convergence after 14 and 9 runs, respectively, which is a reduction of the total number of runs by 40 to 55%. In general, convergence was observed after a lower number of measurement runs for UFP than for PM_{10}. The density plots of the different streets at both study areas was not very different for most of the streets. Only Plantin en Moretuslei in Antwerp generally needed a higher number of repeated runs and Kroonstraat a lower number of runs in comparison to the other streets along the route. In Mol, Statiestraat needed a higher number, and Rozenberg a lower number of repeated runs.

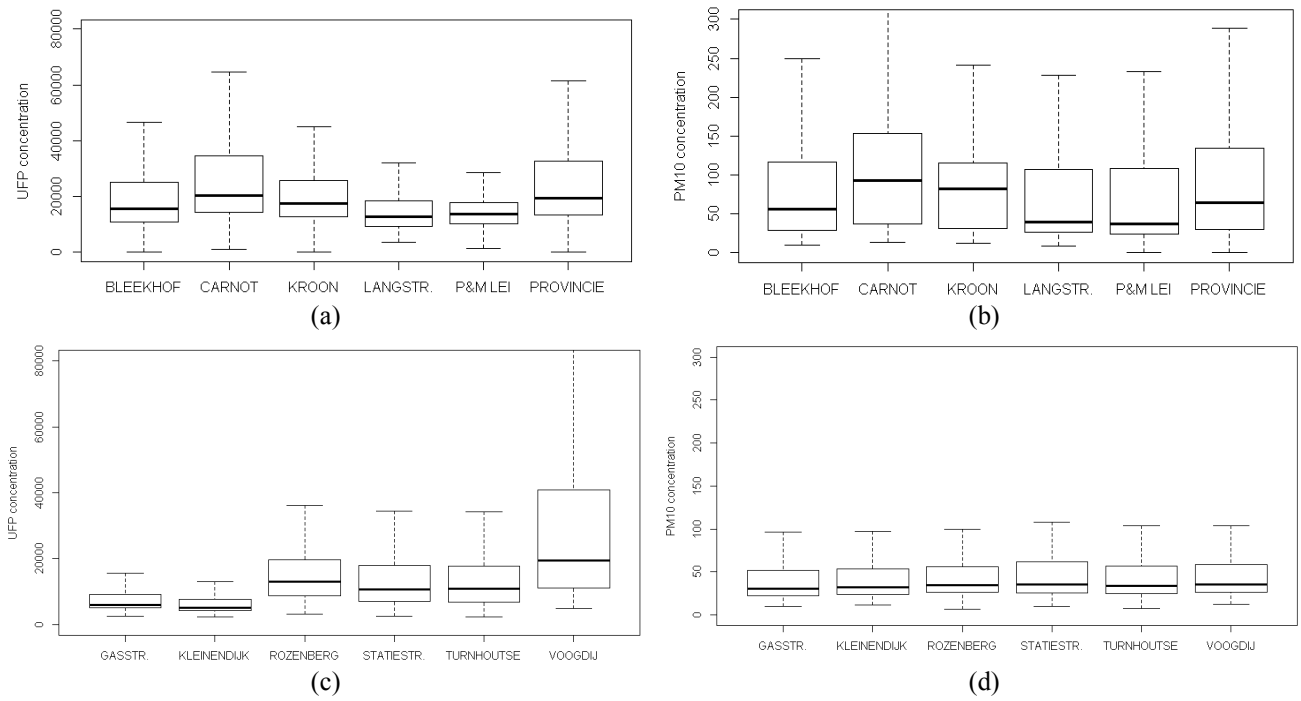

Fig. 5. Boxplots of UFP concentration in Antwerp (a) and Mol (c) and boxplots of PM_{10} concentration in Antwerp (b) and Mol (d) for a selection of streets.

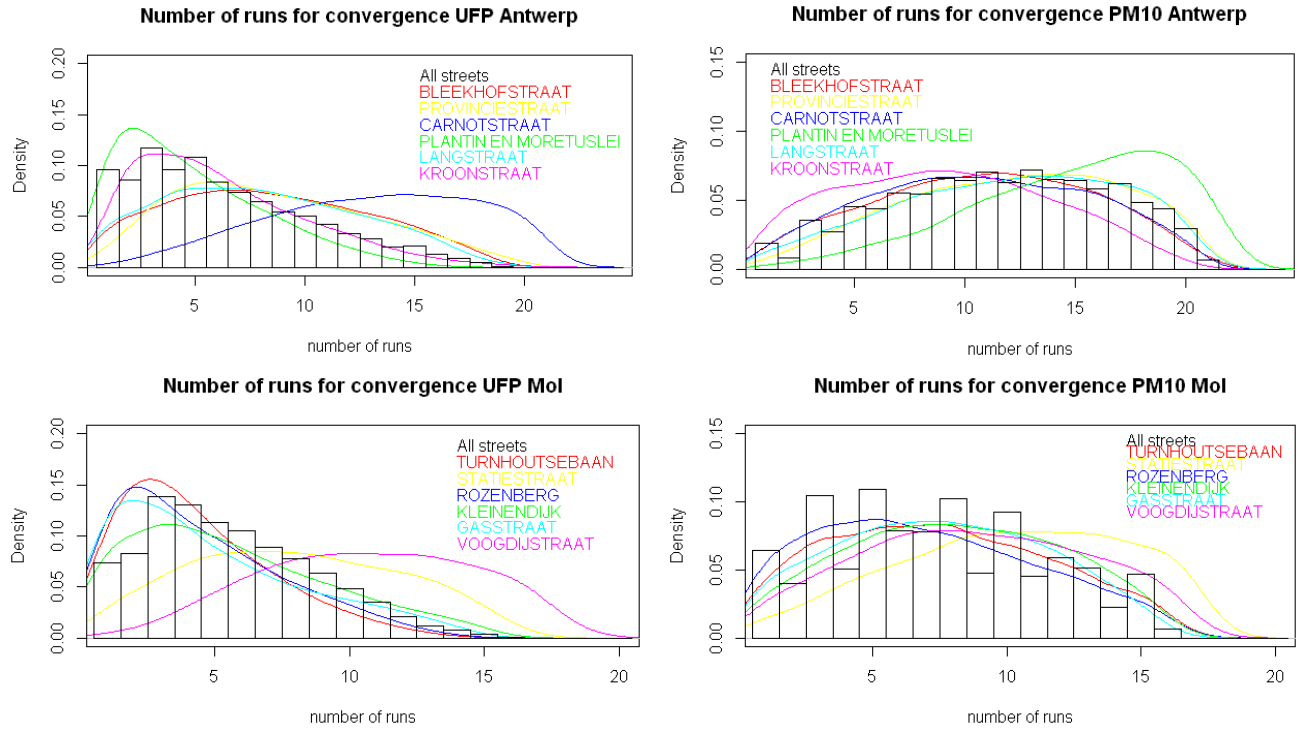

Fig. 6. Plots of the number of runs needed for converge of the median concentration for UFP and PM_{10} at the study sites of Antwerp and Mol. The histogram plot shows the number of runs needed for convergence based on all the measurements, the coloured density plots show the results for specific streets.

An additional qualitative assessment of the temporal distribution of the runs at convergence allowed to draw some conclusions on the preferred temporal spread of data acquisition. For the sequence of the sampled runs with a high similarity in time, convergence was obtained later than for the low similarity sequence for UFP (Fig. 7). These results suggest that for monitoring campaigns covering a substantial part of the day (Antwerp, 6 a.m. until 6 p.m.) monitoring runs

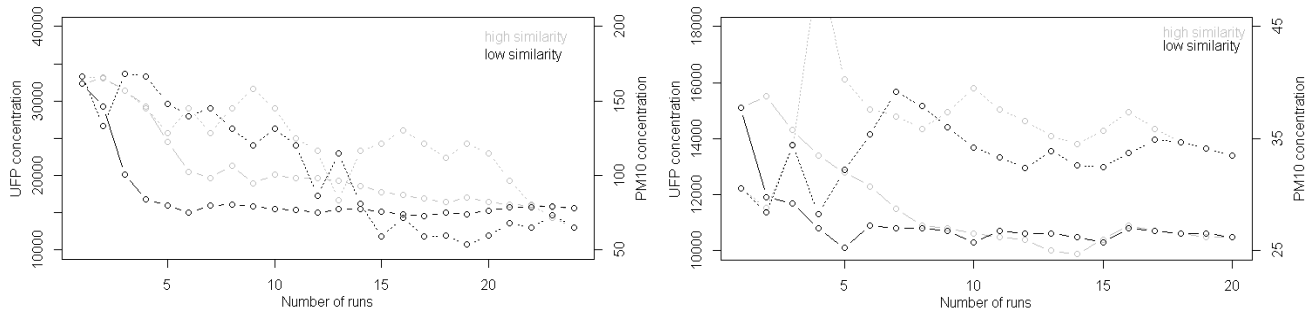

Fig. 7. Median UFP concentration (pt/cm^3, full line) and median PM_{10} concentration (μg/m^3, dotted line) in function of the number of runs, for two contrasting sequences. The sequence with high similarity has strong non-uniform coverage of the measurement period and selected runs made at a similar hour of the day (for Antwerp) or date (for Mol) with higher likelihood. The sequence with low similarity applied an opposite selection, preserving a more uniform coverage.

are better spread over the entire period of the day. When the monitoring campaign is focused over a short period of the day, a temporal spread of the runs over several measurement days also results in a quicker convergence. PM_{10} in Antwerp needed a higher number of repeated runs. In contrast, the sequence of low similarity sequence in Mol contained runs from all the different measurement dates for a cumulative addition of 10 runs. In this specific case, the PM_{10} concentration after 10 runs (34.2 μg/m^3) is very close to the overall median of 33.5 μg/m^3, and 1 run each day would satisfy the data requirements to obtain representative PM_{10} concentrations.

DISCUSSION

Several studies have demonstrated that exposure of people to air pollution is poorly described by central monitoring. Mobile measurements are often presented as complementary to or an alternative for these stationary air quality measurements. They are used in studies on human exposure to quantify personal exposure of individuals and demonstrate the importance of differences in exposure in different micro-environments (Dons *et al*., 2011). However, these studies are still restrained to a limited number of individuals, and do not allow extrapolation to large groups of people.

Only few papers have been published that try to quantify and map air pollution systematically using mobile measurements (e.g., Hagler *et al.*, 2010). Given the spatio-temporal dynamics of the pollutants under investigation and the fact that mobile measurements provide snap-shots of pollutant concentration in space and time, and their sensitivity to short accidental variations and bias (e.g., when the mobile measurement platform is stuck behind a bus in slow moving traffic), the representativeness of the mobile measurements is a major issue. In comparison to stationary measurements spatial resolution is increased at the expense of temporal resolution. Data have to be aggregated both in space and in time in a meaningful way. Finally, mobile measurements can be used to detect hot spots. In this case relative values (comparing different locations and/or different times of the day) are more important than absolute values.

In this study we investigated to what extent a limited set of mobile measurements allows to draw conclusions on urban air pollution and its spatio-temporal variation by using two datasets of mobile measurements. These data sets might be thought of as data sets collected by volunteers that often take the same route but not necessarily every day, neither always at the same moment of the day. The measurement runs are not systematically spread over the days of the total campaign neither over a specific time frame of the day. This results in a sparse data set as it comes to temporal coverage. A crucial question is if these data can be used to draw more general conclusions on the UFP and PM_{10} concentrations in the investigated area. Both an absolute and a comparative perspective can be taken. To try to answer this question we used a pragmatical and empirical approach based upon actual measurement data. First we will report the results of similar measurement campaigns in literature. Then we will take a comparative viewpoint when looking at spatio-temporal variability. Finally, we will discuss the results of the data experiment.

Comparison with Other Measurements

Literature reports on (meta-analyses of) UFP and PM concentrations in relation to the urban micro-environment or transport mode. A comparison of the measurements of this study with the literature is challenging because of potential differences in monitoring equipment or local conditions. The comparison given here should therefore be understood as a qualitative assessment, rather than a fully quantitative comparison of absolute concentrations. The average UFP concentration of all measurements in Antwerp ($2.2 \cdot 10^4$ pt/cm^3) and in Mol ($1.6 \cdot 10^4$ pt/cm^3) is (slightly) higher than the average UFP concentrations for urban environment of $1.08 \cdot 10^4$ pt/cm^3 reported in the meta-analysis of 71 UFP measurement studies by Morawska *et al.* (2008) based on condensation particle counters (CPCs) or differential/scanning mobility particle sizers (DMPS/SMPS). The values are, however, lower than the values reported for street canyons and roadside environments ($4.21 \cdot 10^4$ and $4.82 \cdot 10^4$ pt/cm^3, respectively (Morawska *et al.*, 2008)) due to specific pollutant related processes in these micro-environments. The meta-

analysis of Knibbs *et al.* (2011) based on 599 measurement trips in cycling mode also reports an UFP concentration interval ($3.4 \cdot 10^4 \pm 1.8 \cdot 10^4$ pt/cm^3) in which our measurements fit. The monitoring studies that observed most similar UFP concentrations in urban settings and that were included in this meta-analysis were made in Mol (Bergmans *et al.*, 2009; Int Panis *et al.*, 2010), in Antwerp (Jacobs *et al.*, 2010) and at some locations along a biking path with heterogeneous UFP concentrations in Vancouver (Thai *et al.*, 2007) using TSI P-Trak (Model 8525). In several Dutch cities, Boogaard *et al.* (2009) observed similar values using TSI CPC (Model 3022). In Can *et al.* (2011) an average UFP (< 500 nm, TSI monitor 3031) concentration of $1.2 \cdot 10^4$ pt/cm^3 was reported from stationary measurements on a location nearby the mobile route of Antwerp. UFP concentrations in larger cities and at busy traffic axes are generally higher. Based on P-Trak particle number counts, Kaur (2005) measured an average exposure to UFP for cyclists of 93,968 pt/cm^3 in central London, where concentrations at the backstreets were still as high as 71,628 pt/cm^3, whereas the UFP concentration at high traffic intensity route in Arnhem was 46,600 pt/cm^3 (Zuurbier *et al.*, 2010, using CPCs, model 3007, TSI). These high concentrations were only rarely reached during our measurement campaign at the busiest and more enclosed streets in Antwerp (Carnotstraat and Provinciestraat) and Mol (Voogdijstraat). The UFP concentration measured at the recreational green area in Mol was lower than UFP measurements at park locations in larger cities (e.g., 10,800 pt/cm^3 in Milan, Cattaneo *et al.*, 2009, using CPCs, model 3007, TSI).

Many studies on UFP also report PM_{10} concentrations. Berghmans *et al.* (2009) reported an average PM_{10} concentration of 62.4 in Mol, which is higher than the concentration measured in this study (average of 45 μg/m^3) and may be attributed to the passage at two construction sites during the measurement campaign in the former study. The average PM_{10} concentrations over each daily bicycle traverse in Vancouver ranged between 26 and 77 μg/m^3 (Thai *et al.*, 2009) and were comparable to the concentrations in Mol, but lower than the concentrations measured in Antwerp. Both Berghmans *et al.* (2009) and Thai *et al.* (2009) used GRIMM 1.108 monitor. Overall the UFP and PM_{10} concentrations measured with the mobile platform are in the range of the values reported in the literature for similar biking environments in urban settings.

Comparison of the PM_{10} data to the nearest monitoring stations of the Flemish Environmental Agency (VMM) revealed large differences on some occasions, especially on the occasions with high PM_{10} concentrations. These differences are probably explained by the fact that the VMM station is situated on a backward location at roughly 30 m from the main road in Antwerp, and on a rural site in the case of Mol, i.e., at an (urban) background location, which decreases the contribution of local traffic related sources. The different monitoring technologies used on the mobile platform (optical) and at the reference stations (beta attenuation), is probably causing additional bias. The mobile measurements and the VMM measurements follow the same trend, although peaks are much less pronounced in the VMM data. Therefore, a significant part of the temporal variability is caused by temporal changes in background concentration. A linear model of the median PM_{10} concentration of each measurement run in function of the PM_{10} concentration at (urban) background explained 90% and 80% of the temporal variability of the PM_{10} concentrations for Antwerp and Mol, respectively. A similar comparison could not be made for UFP because UFP is currently not measured by the Flemish Environmental Agency.

Spatio-temporal Variability of Urban UFP and PM_{10} Concentration

Small-scale spatio-temporal gradients in particle concentrations are already well described in the literature (e.g., Monn, 2001, and reference therein). They are caused by the spatio-temporal variability of a multiplicity of sources, dispersion and removal mechanisms (Krudysz *et al.*, 2009). In an urban area, motor vehicle emissions usually constitute the most significant source for UFP (Zhu *et al.*, 2002). Typical background concentrations are far below the urban UFP concentration, and have a very small contribution to the urban UFP concentration (Nikolova *et al.*, 2011). The location and lay-out of roads and the dynamics of the traffic (volumes, speed, fleet composition) are the main factors affecting the spatio-temporal heterogeneity of the UFP concentration. Furthermore, atmospheric dispersion, particle coagulation and particle deposition mechanisms are also influenced by building density, by the geometry of the street and the urban canopy and by meteorology therefore attributing to the spatio-temporal variability of UFP number concentrations. Our measurement results are consistent with these findings in the literature. For UFP significant differences were found between measurement days and between runs on the same day (Fig. 3). It is well known that important diurnal changes occur in UFP concentrations due to changing traffic and other local sources throughout the day (Martin *et al.*, 2009) and these source variations are likely to cause the temporal UFP variations in our study as well.

Lenschow *et al.* (2001) identified long range transport, motor vehicle exhaust and tyre abrasion and resuspension of soil particles as the major PM_{10} sources in urban areas. All these components have their own spatio-temporal dynamics. The contribution of urban background to the PM_{10} at the road, which was observed to be around 60% in Berlin (Lenschow *et al.*, 2001) is more important than the background component in UFP measurements at the roadside due to the short atmospheric residence time of the latter. Our measurements indicate that for PM_{10} the differences between the days are more pronounced whereas intraday variation is often non-significant. The latter does not mean that intraday variability of PM_{10} on these days was non-existent, but may be explained by the importance of the background contribution which masks the short-term local variability of PM_{10}.

Overall, we found a higher small-scale spatial variation for UFP than for PM_{10}. Clayton *et al.* (1993) found a homogeneous spatial distribution of PM_{10} in an urban setting (spatial correlation of 0.93 overnight and 0.9 during

the day). High correlations are also reported for other cities, around 0.8 in Philadelphia and between 0.7 and 0.8 in Los Angeles, but at other locations significant differences up to 20% have been observed between neighbouring locations (Monn, 2001, and reference therein). A small-scale PM_{10} gradient in function of the distance from the road showed the highest spatial variation between measurement locations at 2 m and 15 m from the road (Monn *et al.*, 1997). PM_{10} concentrations further away from the road were comparable due to a good horizontal mixing. A good horizontal mixing of locally emitted PM_{10} along the routes in this study is likely to explain the limited spatial variation observed between the streets.

To allow for a systematic comparison between different locations all data were aggregated at street level. This streetwise comparison shows significant differences for UFP between streets both in Antwerp and in Mol. For PM_{10} the spatial variability is lower. The results suggest that the added value of mobile measurements for mapping spatial variability of air quality is more important for UFP than for PM_{10}. In Mol, streets with higher traffic volumes and a canyon configuration (e.g., Voogdijstraat) could clearly be distinguished from more quiet backend (Kleinendijk) or more open streets (Turnhoutsebaan). This is also apparent in Antwerp, where UFP concentrations in the street with the highest traffic volume (Plantin en Moretuslei) were surprisingly comparable to the streets with the lowest traffic volume (Langstraat), and significantly lower than streets with intermediate traffic volumes (Carnotstraat and Provinciestraat). This is probably caused by the street lay-out with a separate biking lane at several metres distance from the traffic lanes and rather smooth traffic, whereas in Carnotstraat and Provinciestraat cyclists ride right next to or even in the wake of the cars and traffic gets easily congested. This street-level aggregation does not take into account intra-street variability. On the other hand this allows to level out part of the variability that is related to traffic discontinuity and short-term incidents. We assume most streets that were included in the measurement campaigns, to be discontinuous line sources with rather homogenous lay-out.

Data from runs on different days and on different moments of the day are aggregated. A potential source of bias in this streetwise comparison is the fact that diurnal traffic patterns might be different between streets, and that sampling could mainly have taken place at moments corresponding to high traffic in one street (e.g., end of school) and simultaneous low traffic in other streets (e.g., not affected by school traffic). Both issues can be tackled by increasing the number of runs. This would allow to aggregate the data at a more detailed level, i.e., distinguishing non-homogenous street sections and intersections, and aggregating data in relevant time intervals, e.g., hourly averages, peak and off-peak hours, etc.

To find out how sensitive the results are for the number of runs and for the timing of the runs, we carried out an experiment on the data. In the streetwise comparison we use the median pollutant concentration per street. However, we don't have an absolute reference to compare these concentrations to. If after progressively adding runs and calculating the median concentration for these runs, the median converges quite quickly and consistently to the overall median concentration, we have a strong indication that the overall median concentration is representative for the considered period. However, this median value can be affected by the fact that no runs were carried out on days with overall substantially higher or lower concentrations. The results showed that the moment that convergence is obtained for UFP differs from 1 to 18 or 16 runs for Antwerp and Mol, respectively. In fact, a substantial reduction of 70% of the number of runs would still be enough to obtain a reasonable estimate of the overall median concentration, but less so for streets with higher traffic density and a canyon like configuration. E.g., in Mol, the streets with moderate traffic and street canyon configuration showed the highest number of runs needed for convergence. The street with the highest traffic density of this study (Plantin en Moretuslei), however, showed the fastest convergence for UFP, probably due to the separate biking lane and the consistently high traffic counts throughout the day. Its convergence pattern was comparable with the pattern found for Kleinendijk in Mol, which is a recreational and green area with very limited local sources. Therefore an a priori definition of a suitable number of runs based on traffic density is hard to make. However, the results indicate that less runs would be needed for streets with low or smooth traffic, whereas more runs are needed in streets with regular periods of congestion. Irrespective of the street and traffic characteristics, the results suggest that it is better to have a spread of the measurement runs over the considered daytime interval and over the entire measurement period, than to have them concentrated. For PM_{10} the moment that convergence is obtained is generally higher, and the reduction of the number of runs lower (40% in Antwerp, 55% in Mol). This can be explained by the fact that the temporal variability of PM_{10} at the street is determined by the variability of the local sources but mainly by the variability of the background concentration. In Mol, a median PM_{10} concentration close to the overall median could be obtained after 10 runs, given that these runs were made on the 10 different dates of the measurement campaign.

Our results indicate that the use of a limited set of about 20 mobile measurements carried out on different days and different times of the day allows to distinguish streets with higher and lower median concentrations of UFP in a significant way. Strictly speaking these relative differences are only valid for the period of sampling, but assuming traffic patterns in all streets are similarly affected by seasonal variations or holiday periods, these differences are indicative for the whole year.

CONCLUSION

This paper illustrates the potential of mobile measurement methods to map urban air pollution both for comparative purposes and for estimation of absolute levels of UFP and PM_{10}. We demonstrated that a limited number of 20 to 24 runs carried out on different days and different times of the day over a period of two to three weeks allows to distinguish

streets with higher and lower median concentrations of PM_{10} and UFP in a significant way. We also presented strong indications that the same set of measurement runs provides a good quantitative approximation of median UFP and PM_{10} concentrations for most streets, and that this number could even be reduced. We conclude that a limited set of mobile measurements can be used to detect hot spots and locations with systematically higher or systematically lower UFP and PM_{10} levels in an urban environment. Mobile measurements are thus well suited to be used in an opportunistic monitoring scheme, in which volunteers (e.g., postmen) carry measurement equipment with them in their normal daily routine. In this way both spatial and temporal coverage can be further increased, and the measurement period can be extended. When used for comparative purposes further improvement is possible by rescaling the data to take into account day to day variation in background concentrations.

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