Atmospheric Environment 112 (2015) 306–316

Contents lists available at ScienceDirect

Atmospheric Environment

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

Identifying drivers for the intra-urban spatial variability of airborne particulate matter components and their interrelationships

Hao Wu [a, b, *], Stefan Reis [b, c], Chun Lin [a], Iain J. Beverland [d], Mathew R. Heal [a]

[a] University of Edinburgh, School of Chemistry, Joseph Black Building, David Brewster Road, Edinburgh, EH9 3FJ, UK
[b] Centre of Ecology & Hydrology, Bush Estate, Penicuik, Edinburgh, Midlothian, EH26 0QB, UK
[c] University of Exeter Medical School, Knowledge Spa, Truro, TR1 3HD, UK
[d] University of Strathclyde, Department of Civil and Environmental Engineering, James Weir Building, 75 Montrose Street, Glasgow, G1 1XJ, UK

HIGHLIGHTS

- Peripatetic measurements of UFP, BC and $PM_{2.5}$ were taken in an urban environment.
- Spatial variability in UFP and BC were much larger than in $PM_{2.5}$.
- UFP and BC were significantly correlated with traffic counts, while $PM_{2.5}$ was not.
- $PM_{2.5}$ variability was largely determined by synoptic meteorological influences.

ARTICLE INFO

Article history:
Received 19 January 2015
Received in revised form
21 April 2015
Accepted 24 April 2015
Available online 25 April 2015

Keywords:
Black carbon
Ultrafine particles
$PM_{2.5}$
Spatiotemporal variability
Mobile measurements

ABSTRACT

The aim of this work was to compare the variability in an urban area of fine particles ($PM_{2.5}$), ultrafine particles (UFP) and black carbon (BC) and to evaluate the relationship between each particle metric and potential factors (local traffic, street topography and synoptic meteorology) contributing to the variability. Concentrations of the three particle metrics were quantified using portable monitors through a combination of mobile and static measurements in the city of Edinburgh, UK. The spatial variability of UFP and BC was large, of similar magnitude and about 3 times higher than the spatial variability of $PM_{0.5\text{-}2.5}$ (the PM size fraction actually quantified in this work). Highest inter-daily variability was observed for $PM_{0.5\text{-}2.5}$, which was approximately 2 times higher than inter-daily variability of BC and UFP. Elevated concentrations of UFP and BC were observed along streets with high traffic volumes whereas $PM_{0.5\text{-}2.5}$ showed less variation between streets and a footpath without road traffic. Both BC and UFP were significantly correlated with traffic counts, while no significant correlation between $PM_{0.5\text{-}2.5}$ and traffic counts was observed. BC was significantly correlated with UFP, with significantly different regression slopes between working days and non-working days implying that the increased number of diesel powered heavy goods vehicles during working days contributed more to BC than to UFP. It is concluded that variations in BC and UFP concentrations were mainly determined by the nearby traffic count and varying background concentrations between days, while variation in $PM_{0.5\text{-}2.5}$ concentration was mainly associated with regional sources. These findings imply the need for different policies for managing human exposure to these different particle components: control of much BC and UFP appears to be manageable at local scale by restricting traffic emissions; however, abatement of $PM_{2.5}$ requires a more strategic approach, in cooperation with other regions and countries on emissions control to curb long-range transport of $PM_{2.5}$ precursors.

1. Introduction

Evidence continues to accumulate of the adverse health impacts of $PM_{2.5}$, the mass concentration of airborne particulate matter (PM) with an aerodynamic diameter of less than 2.5 μm (WHO, 2013). However, metrics of other characteristics of ambient PM

* Corresponding author. University of Edinburgh, School of Chemistry, Joseph Black Building, David Brewster Road, Edinburgh, EH9 3FJ, UK.
E-mail address: h.wu@ed.ac.uk (H. Wu).

http://dx.doi.org/10.1016/j.atmosenv.2015.04.059

including numbers of ultrafine particles (UFP, particles of diameter <100 nm) and black carbon (BC) concentrations are emerging as important in terms of their association with health effects (Heal et al., 2012). A relevant issue is the extent to which UFP and BC concentrations vary within populated areas, since a shortcoming in many epidemiological studies is assumption of homogenous exposure within the study area. This might be plausible for pollutants with less spatial variability but could result in significant bias in exposure-response relationships for highly spatially variable pollutants (Hoek et al., 2002). In this context, variables related to the contribution of emissions from major roads (e.g. traffic intensity, or distance to the road) are commonly identified as significant predictors for a range of traffic-related air pollutants in many studies applying land-use regression models (Hoek et al., 2008), the validity of which might be influenced by the underlying causes of the variability of different pollutants. Thus one of the aims of this work was to evaluate the extent to which potential factors affect the spatiotemporal variability of ambient BC, UFP and $PM_{2.5}$ in an urban area. These factors include local traffic, street topography and synoptic meteorology which, although recognised in the literature, have rarely been compared in terms of their influences on different metrics.

The three airborne particle metrics are closely related to traffic in urban environments (HEI, 2010; Kassomenos et al., 2014; Sandradewi et al., 2008) but, for $PM_{2.5}$ in particular, synoptic-scale meteorology also affects the dispersion and long-range transport of secondary particles (Pinto et al., 2004). UFP variability is also subject to high intensity secondary formation associated with strong solar radiation (Reche et al., 2011). Street canyons, which are ubiquitous in many urban environments, introduce complex dispersion characteristics that further increase the spatial variability of BC and UFP (Peters et al., 2014; Rakowska et al., 2014). One limitation of inter- and intra-urban studies of airborne particle concentrations is that the fixed-site measurements on which they are based are rarely sufficient in number, and thus in spatial coverage, to explore the variability of exposure to particles at street level. One way to monitor particle concentration at high spatiotemporal resolution is by use of mobile monitoring instruments, which has good prospects for wider application in the assessment of human exposure to air pollution in the future (Steinle et al., 2013).

Both UFP and BC have received increasing interest in recent studies (Patton et al., 2014; Ruths et al., 2014), as they can be considered markers of a range of traffic-related particulate pollutants. Therefore the relationships of UFP and BC with traffic volume and composition need to be understood in order to correctly assign exposure to traffic pollution in health studies. In the UK and many other countries, $PM_{2.5}$ and PM_{10} are the only two regulated PM metrics (AQEG, 2012). Given the increasing evidence for the harmfulness of UFP (WHO, 2013) with its ability to penetrate deep into the airways (Knibbs et al., 2011), investigation on the relationship between UFP and $PM_{2.5}$ can provide insight on the extent to which current policy can effectively protect human health. Thus another aim of this work was to investigate the inter-relationships between the different metrics of PM and their relationships with traffic.

In this work, pairs of portable instruments were used to measure $PM_{0.5\text{-}2.5}$ (used here as a measure of $PM_{2.5}$), UFP number and BC concentrations within the city of Edinburgh (Scotland) in two series of measurement campaigns in winter and in spring. Analyses of data from a combination of mobile and stationary measurements were used to evaluate possible causes of the variations in the concentrations of the different PM metrics.

2. Methods

2.1. Study design

BC, UFP and $PM_{0.5\text{-}2.5}$ concentrations were measured across the south of the city of Edinburgh, UK (55.9° N, 3.2° W, population ~480,000) in two separate campaigns using 2 units of the following instruments: microAeth AE51 (AE51), TSI 3007 Condensation Particle Counter (CPC) and Dylos Corp. DC1700 (Dylos).

In the winter campaign, between December 2013 and January 2014, the measurements were conducted three times on Mondays and once on Sunday primarily near roadside by walking between and pausing at designated sites (Fig. 1a). The sites were selected to cover potential hotspots, urban background sites (at least 130 m from the nearest major road) and different street topographies

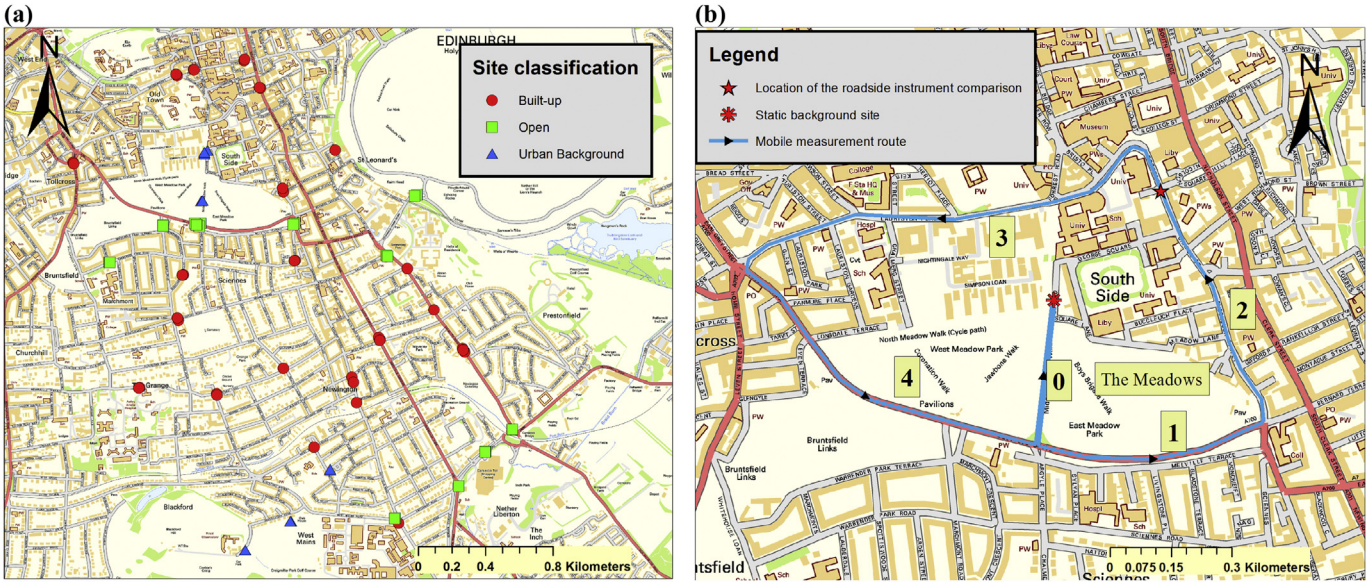

Fig. 1. (a) Location and classification of the static measurement sites. Streets with buildings on both sides are classified as built-up. Streets with buildings on only one side or no buildings on either side are classified as open. Background sites are at least 130 m away from the nearest major road. (b) Mobile measurement route and location of the contemporaneous background measurements. Segments of the mobile route are labelled from 0 to 4. Base map from Edina Digimap®.

(open or built-up) over an area of about 6 km^2. In a typical walk, the measurements started at around 10:00 a.m. and proceeded through the designated sites to the final location roughly in the order from south to north and from east to west (Fig. 1a). At each site a 5-min static measurement was conducted, during which the number and type of vehicles (car, van, heavy goods vehicle and bus) passing the observer were recorded. Throughout each walk, measurements for each pollutant were taken in parallel (with duplicate instruments) on both sides of the road. To evaluate the duplicate precision, inter-comparison between the pairs of instruments was conducted in a separate trial on Mon 3rd Feb 2014 from 10:00 a.m. to 12:00 p.m. by walking through the same route but with the duplicate instruments carried by one person. The weather conditions on this day were similar to other measurement days (Supplementary Information Table S1). Traffic characteristics are assumed to be similar to other Monday measurements.

In the spring campaign, between April and May 2014, measurements were taken around a park area (~1 km^2), referred to locally as 'the Meadows', focusing on understanding the contributions from traffic-related sources and local background sources to BC, UFP and $PM_{0.5\text{-}2.5}$ on typical urban streets (Fig. 1b). The road on the south edge of the Meadows had an annual average daily flow of 13,272 vehicles in 2013 (Dft, 2015). The pollution level associated with traffic was monitored by walking along a route surrounding the Meadows. To measure temporal variation in the background concentrations, a duplicate set of instruments was located at a static site inside the route circuit during the collection of the mobile measurements. This background location had perpendicular distance between 160 and 480 m to the three sides of the triangle route (Fig. 1b). The measurements were conducted on one Sunday and five weekdays. On each day the mobile measurements started together with the static measurements at the background site and proceeded in the directions indicated in Fig. 1b. Two trips were carried out in the morning (~9–10 a.m.) and early afternoon (~1–2 p.m.) during each day, except for adverse weather conditions on one of the weekday afternoons. Other incomplete sets of measurements during each day were due to instrument faults. The route of the mobile measurements was divided into five segments with different street topographies and traffic densities, as labelled on Fig. 1b. The total traffic passing the observer in the direction of the route was counted for each segment. Traffic flow in the opposite direction of the route is assumed to be similar. The duplicate instruments were compared against each other during the last four measurement days by co-location for at least 20 min either at the static background site or near a busy roadside (Fig. 1b). The inclusion of both background and roadside sites was to cover a range of concentrations for the evaluation of duplicate instrument precision.

2.2. Instrumentation

The AE51 determines BC concentration from absorption of 880 nm laser light by particles continuously collected on a glass-fibre filter. The CPC measures particle number concentrations of particles between 0.01 and 1 μm in diameter by using laser light scattering after condensing particles with super-saturated iso-propanol vapour. Although UFP is usually defined as particles smaller than 100 nm, since the number concentration is dominated by ultrafine particles the measurement from the CPC can be considered to represent UFP number concentration. The Dylos measures the particle number concentration using laser light scattering technique in two size ranges, >0.5 and >2.5 μm. Only particles between 0.5 and 2.5 μm in diameter were included in this study and are thus referred to as $PM_{0.5\text{-}2.5}$. The use of the terminology $PM_{2.5}$ elsewhere in this paper refers to the mass of all particles <2.5 μm as defined for air quality standards.

In the winter campaign the time bases for the AE51, CPC and Dylos were 1 min, 1 s and 1 min, respectively. Because of the shorter duration of a trip in the spring campaign the resolution for the AE51 was increased to 30 s to ensure sufficient data points for each segment. The Optimised Noise-reduction Averaging (ONA) algorithm was used to reduce the noise in the data recorded by the AE51 (Hagler et al., 2011). The ONA algorithm conducts adaptive time-averaging of the BC data, with the incremental light attenuation (ΔATN) through the instrument's internal filter determining the time window of averaging. The ΔATN thresholds were set at 0.01 and 0.05 for winter and spring measurements, respectively, as a result of the different proportions of clean background areas and sampling resolutions in two campaigns. Negative values recorded after the smoothing were omitted from further analyses (consisting of ~5% of the whole data set), which mostly occurred when the measured concentrations were <100 ng/m^3.

Major axis (MA) regression analysis was carried out to test the equivalence between duplicate instruments, assuming that the uncertainties in the duplicate instruments are similar (Warton et al., 2006). A statistical summary of instrument inter-comparison results is given in SI Table S2. Correlations between duplicate instruments were highly significant, however the 95% confidence interval of the slopes for AE51 and CPC in spring campaign and Dylos in both campaigns did not encompass unity. Therefore corrections based on the slope and intercept from the MA regression analyses were applied to the corresponding instruments to allow comparison between duplicate instruments.

2.3. Additional data

The average $PM_{2.5}$ concentrations measured by a TEOM-FDMS instrument at the UK national network monitoring station St. Leonards (55.945589° N, 3.182186° W) located 600 m to the east of the Meadows during each set of measurements are summarised in SI Fig. S1. The TEOM-FDMS is a reference-equivalent instrument for quantifying gravimetric $PM_{2.5}$ for statutory purposes. Meteorological data for the period of measurement for each day in both campaigns and for the period when instrument inter-comparisons were carried out in the winter campaign were obtained from a weather station on the rooftop of a seven storey building located ~3 km to the south of the Meadows (55.92° N, 3.17° W) and are summarised in Table S1.

2.4. Data analyses

Reduced major axis (RMA) regression analysis was used to investigate the correlation between different pollutants, since the magnitudes of the data values and their uncertainties are not the same for different instruments (Ayers, 2001). Given the skewed nature of pollutant distributions, the non-parametric Mann–Whitney U test and the Kruskal–Wallis test were used to determine whether median concentrations differed significantly between two and more than two samples, respectively. Data analyses were performed mainly using R software (R Core Team, 2014) and sometimes Microsoft Excel. Back trajectory data was imported from pre-calculated trajectory data using the HYSPLIT trajectory model via the "importTraj" function in openair (Carslaw and Ropkins, 2012), an R package for air quality data analysis.

3. Results and discussion

3.1. Spatiotemporal variability of BC, UFP and $PM_{0.5\text{-}2.5}$

Distributions of BC, UFP and $PM_{0.5\text{-}2.5}$ concentrations in the winter measurements are summarised in Fig. 2. The distributions of

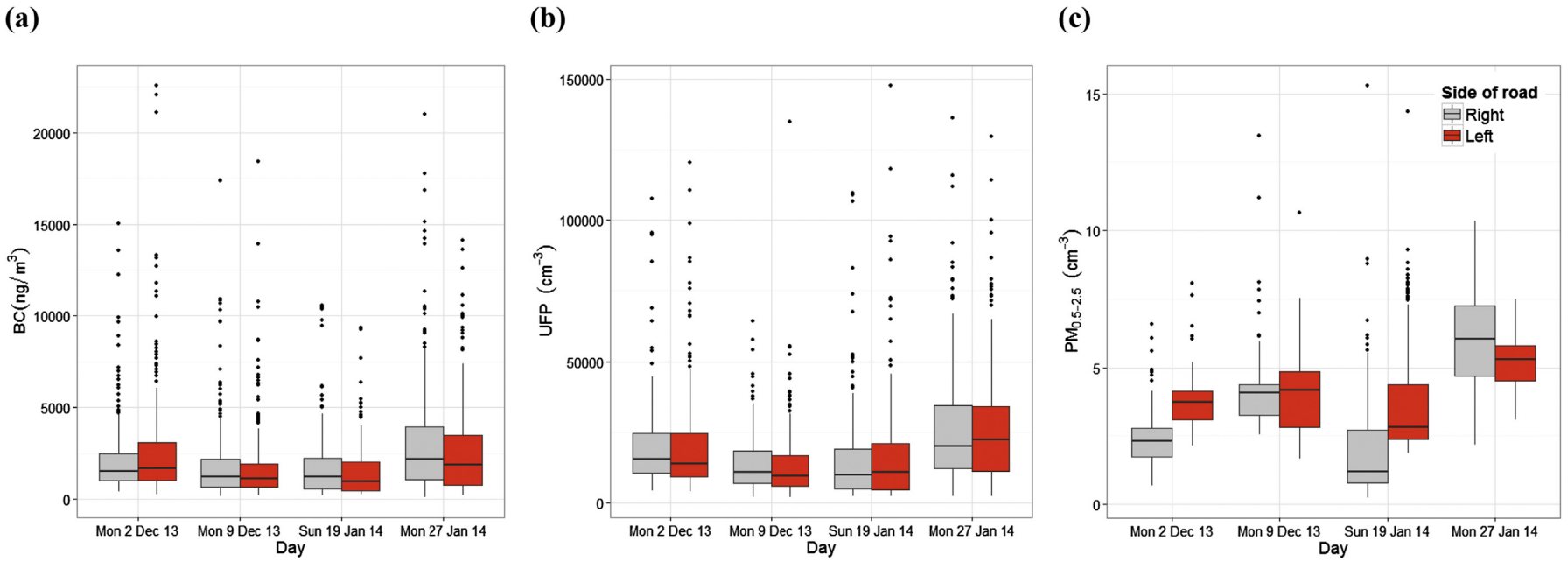

Fig. 2. Distributions of (a) BC, (b) UFP and (c) $PM_{0.5\text{-}2.5}$ concentrations measured on both sides of the road during each week in the winter campaign. The bold horizontal line denotes the median, and the box demarcates the interquartile range. The whiskers extend to the values 1.5 times the IQR on each side of the median. The $PM_{0.5\text{-}2.5}$ concentration measured by one of the Dylos instruments was corrected based on the statistics from MA regression analysis of instrument co-deployment during the winter campaign (Table S2). Side of road is defined with respect to the walking direction in the mobile measurements. The UFP concentrations in (b) are 1 min averages of the raw 1 s data.

pollutant concentrations are highly skewed, especially for BC and UFP, so the interquartile range (IQR) is used to illustrate the variation in distribution. Statistical summaries of the median and IQR for measurements on both sides of the road are listed in Table 1a. BC and UFP concentrations in spring campaign and $PM_{0.5\text{-}2.5}$ concentrations in both campaigns were corrected according to the MA regression analyses results in Table S2 to allow comparison between duplicate instruments. The ratio between IQR and median is used here as a metric of the spatiotemporal variability of each pollutant during each measurement trip. Table 1a shows that BC had the highest variability in the winter campaign (average IQR/median ratio of 1.34), which is about twice as high as for the metric with lowest variability, $PM_{0.5\text{-}2.5}$ (average IQR/median ratio of 0.56). The difference in concentrations measured on two sides of the road was not significant for BC and UFP as the median on one side of the road was always within the IQR of the other side (Fig. 2). $PM_{0.5\text{-}2.5}$

Table 1
The median, IQR and ratio between IQR and median on each day for BC, UFP and $PM_{0.5\text{-}2.5}$ for (a) winter and (b) summer campaign. Left and right side of the road is defined with respect to the walking direction in the mobile measurements. Adjustment derived from MA regression analyses of instrument co-deployments (Table S1) was applied to one set of $PM_{0.5\text{-}2.5}$ data in both campaigns, and one set of BC and UFP data in the spring campaign.

(a) Winter campaign

BC

Date	Right			Left		
	Median (ng/m^3)	IQR (ng/m^3)	IQR/Median	Median (ng/m^3)	IQR (ng/m^3)	IQR/Median
Mon 2nd Dec 13	1481	1540	1.04	1668	2112	1.27
Mon 9th Dec 13	1210	1521	1.26	1105	1336	1.21
Sun 19th Jan 14	1195	1664	1.39	951	1571	1.65
Mon 27th Jan 14	2155	2900	1.35	1842	2834	1.54

UFP

Date	Right			Left		
	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median
Mon 2nd Dec 13	15,251	14,132	0.93	13,971	15,870	1.14
Mon 9th Dec 13	10,664	11,412	1.07	9481	10,759	1.13
Sun 19th Jan 14	9649	14,030	1.45	10,916	16,715	1.53
Mon 27th Jan 14	19,890	22,349	1.12	22,502	23,490	1.04

$PM_{0.5\text{-}2.5}$

Date	Right			Left		
	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median
Mon 2nd Dec 13	2.30	1.07	0.46	3.74	1.04	0.28
Mon 9th Dec 13	4.06	1.13	0.28	4.18	2.05	0.49
Sun 19th Jan 14	1.18	1.91	1.62	2.80	1.98	0.71
Mon 27th Jan 14	6.05	2.60	0.43	5.29	1.29	0.24

(b) Spring campaign

BC

Date	Mobile			Static		
	Median (ng/m^3)	IQR (ng/m^3)	IQR/Median	Median (ng/m^3)	IQR (ng/m^3)	IQR/Median
Sun 6th Apr 14	251	282	1.12	23	3	0.14
Thu 10th Apr 14	955	1027	1.08	276	470	1.70
Fri 18th Apr 14	1035	654	0.63	540	391	0.72
Wed 23rd Apr 14	2124	2208	1.04	1779	594	0.33
Wed 30th Apr 14	1672	1850	1.11	1154	288	0.25
Wed 7th May 14	926	1208	1.30	217	635	2.92

UFP

Date	Mobile			Static		
	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median	Median (cm^{-3})	IQR cm^{-3})	IQR/Median
Sun 6th Apr 14	1757	1891	1.08	624	509	0.82
Thu 10th Apr 14	10,276	8824	0.86	6081	6560	1.08
Fri 18th Apr 14	38,009	24,034	0.63	30,128	22,487	0.75
Wed 23rd Apr 14	11,370	8674	0.76	16,115	15,642	0.97
Wed 30th Apr 14	9633	17,816	1.85	4547	2172	0.48
Wed 7th May 14	6431	8353	1.30	3286	866	0.26

$PM_{0.5\text{-}2.5}$

Date	Mobile			Static		
	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median	Median (cm^{-3})	IQR (cm^{-3})	IQR/Median
Sun 6th Apr 14	0.61	0.42	0.69	1.87	0.21	0.11
Thu 10th Apr 14	3.70	0.51	0.14	3.46	0.22	0.06
Fri 18th Apr 14	4.64	0.47	0.10	4.41	0.43	0.10
Wed 23rd Apr 14	19.46	3.67	0.19	18.23	5.99	0.33
Wed 30th Apr 14	15.31	3.49	0.23	16.42	3.05	0.19
Wed 7th May 14	2.00	0.75	0.38	2.49	0.44	0.18

showed noticeable difference in distributions between two sides of the road. However this may have resulted from the use of a single linear equation to correct the Dylos data. A 3-day comparison between the duplicate Dylos instruments in an urban background environment (55.92° N, 3.18° W) at the beginning of the winter campaign (3rd – 5th Dec 2013) showed a significantly different slope from that derived from the inter-comparison at the end of the winter campaign (3rd Feb 2014) (Table S2). Fig. S2 suggests that the relationship between the duplicate Dylos instruments might follow a different linear relationship between different days as indicated by the deviation of scatter points on one day from the main trend line. Due to lack of inter-comparison between the Dylos instruments during each measurement day, it is difficult to quantify the genuine difference in $PM_{0.5\text{-}2.5}$ concentrations between the two sides of road in Fig. 2c. The discrepancies in the relationship between duplicate CPC and AE51 units on different days were considered to be less significant than the Dylos, since the correlation coefficients between CPC and AE51 units ($r > 0.97$) in the mobile winter inter-comparison were much higher and the gradients (0.98 and 0.97, respectively) were much closer to unity than the Dylos equivalents ($r = 0.90$, gradient = 0.44) (Table S2).

Distributions of BC, UFP and $PM_{0.5\text{-}2.5}$ concentrations in the spring measurements are summarised in Fig. 3. Jittered points are plotted for UFP to reveal the overlapping of large numbers of data points. In order to discern the details of the boxes in Fig. 3a and b, BC and UFP concentrations greater than 15,000 ng/m^3 and 100,000 cm^{-3}, respectively, are not included. However, extremely high concentrations were observed, extending to 50,000 ng/m^3 for BC and 400,000 cm^{-3} for UFP. Elevated BC concentrations were observed on streets with traffic (route segments 1–4) compared with the footpath through an area of urban park (route segment 0). The median concentrations for each route segment also varied between different times of a day. Only on Thu 10th Apr 2014 were BC concentrations not significantly different between morning and noon sessions (Mann–Whitney U test). BC concentration on Sun 6th Apr 2014 (median = 251 ng/m^3) was significantly lower than on weekdays (median = 1281 ng/m^3) (one-tailed Mann–Whitney U test, $P < 0.01$), which is similar to findings in the winter

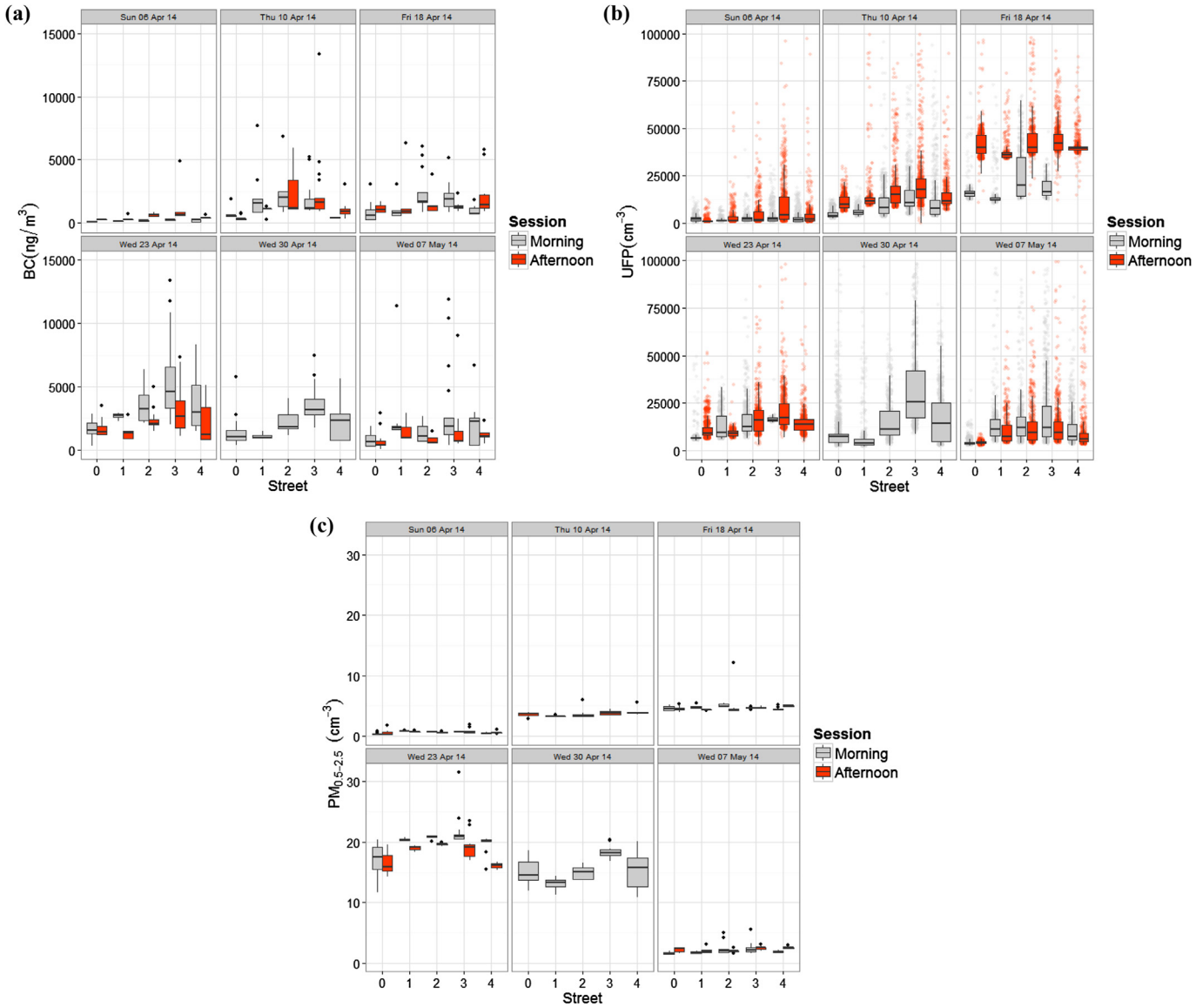

Fig. 3. Box plots for (a) BC, (b) UFP and (c) $PM_{2.5}$ concentrations grouped by different days and sessions in the spring campaign. Data visualisations as defined in Fig. 2. Additional jitter points are plotted in Fig. 2b to reveal the extent of data in the outliers.

measurements.

The large range of outliers in the UFP distributions (Fig. 3b) implied that UFP was greatly influenced by the emissions of nearby traffic. Despite the fact that the outlier measurements on each street were usually a factor 3 higher than their median, the difference in the medians between streets was relatively small, suggesting that elevated levels due to occurrence of exhaust plumes only transiently affected the UFP concentration. Using ratios between median concentrations on streets and on the footpath it was calculated that, on average, a pedestrian was exposed to 1.8 times higher concentration on streets than on the footpath for UFP, and 2.3 times higher for BC. The markedly high UFP level on Fri 18th Apr 2014 at noon was associated with the lowest wind speed, highest solar flux (Table S1) and relatively high O_3 concentration of ~70 μg/m^3 (recorded at the St. Leonards UK national network monitoring station) compared to other days. All these conditions are likely to have promoted secondary particle formation from photochemical reactions (Reche et al., 2011).

The distribution of $PM_{0.5\text{-}2.5}$ concentrations (Fig. 3c) exhibited a different pattern compared with the BC and UFP concentrations. The range of concentration on each street was smaller, and the relative variation in medians between streets within the same day was lower. Only on Sunday and Wednesday 1 were $PM_{0.5\text{-}2.5}$ concentrations highly significantly different between footpath and the streets (Mann–Whitney U test, $P < 0.01$), indicating that local traffic is frequently not a dominant contributor to $PM_{0.5\text{-}2.5}$ concentrations in the locations where measurements were made. Possible traffic-related sources to PM in this size range would be tyre and brake wear and road abrasion, but these are considered to contribute relatively little to $PM_{2.5}$ (AQEG, 2012). Significant variation in $PM_{0.5\text{-}2.5}$ concentrations was noticed between all spring working sampling days (Kruskal–Wallis test, $\chi^2 = 297$, df = 3, $P < 0.01$), which implies that $PM_{0.5\text{-}2.5}$ is more influenced by the regional sources and meteorological conditions on a particular day rather than the local traffic emissions. This is consistent with observations in other cities, e.g. relatively low contribution (13%) to $PM_{2.5}$ from local traffic was also found in Paris (Skyllakou et al., 2014). Despite the Dylos not measuring particles smaller than 0.5 μm, $PM_{0.5\text{-}2.5}$ variation between days was in very good relative agreement with that of the $PM_{2.5}$ concentration measured by TEOM-FDMS at the St Leonard's national network urban background site (SI Fig. S1), i.e. highest $PM_{2.5}$ levels on Mon 27th Jan 2014 in winter, and on Wed 23rd Apr 2014 and Wed 30th Apr 2014 in spring, and lower on Sundays in both campaigns.

Assuming that UFP represents contribution from local traffic sources, the ratio between median UFP and $PM_{0.5\text{-}2.5}$ approximates the relative contribution of local sources to $PM_{2.5}$ concentration during that day. However caution should be made in this interpretation since any photochemical new particle formation (e.g. Fri 18th Apr 2014 in spring campaign) may lead to overestimation of the relative level of local contribution compared to days when such an event is unfavourable. In the winter measurements, the highest UFP/$PM_{0.5\text{-}2.5}$ ratio (3999), was associated with relatively low $PM_{2.5}$ concentration (8 μg/m^3) on Sunday (Fig. 4). In the spring measurements, the highest ratio was on Fri 18th Apr 2014 (3715), which was about ten times the ratio on Wed 23rd Apr 2014 (349), despite the fact the $PM_{2.5}$ concentration on Fri 18th Apr 2014 (~9 μg/m^3) was only a third of that on Wed 23rd Apr 2014 (~27 μg/m^3). It is noted that in both campaigns highest local contributions did not coincide with the highest $PM_{2.5}$ concentrations. This observation again indicates a strong regional component in $PM_{2.5}$ observed in urban areas, as also noted by AQEG (2012).

Four-day air-mass back trajectories arriving at Edinburgh at 09:00 a.m. and 12:00 p.m. on each measurement day are plotted in SI Fig. S3. The trajectories associated with the highest $PM_{2.5}$ concentrations in the winter and spring campaigns originated from North America and northern continental Europe, respectively. However, the highest $PM_{2.5}$ concentration in winter coincided with a relatively large contribution from local sources (UFP/$PM_{0.5\text{-}2.5}$ ratio of 2682) suggesting that the high $PM_{2.5}$ on that particular day may be mainly due to local emission sources. In contrast, the south-easterly trajectory originating from northern continental Europe (Fig. S3) was associated with high $PM_{2.5}$ concentrations and low local contributions. It is notable that the $PM_{2.5}$ concentrations on the days associated with relatively high local emissions were close to the Scottish annual air quality objective threshold concentration for $PM_{2.5}$ of 12 μg/m^3 to be achieved by 2020 (AQEG, 2012), and that the two days with $PM_{2.5}$ concentrations breaching the limit (Weds 23rd and 30th Apr 2014 in spring, Fig. S1) had the largest regional contribution. In the UK context, transboundary import of inorganic aerosol components resulting in high particle concentrations was reported in a modelling study (Vieno et al., 2014).

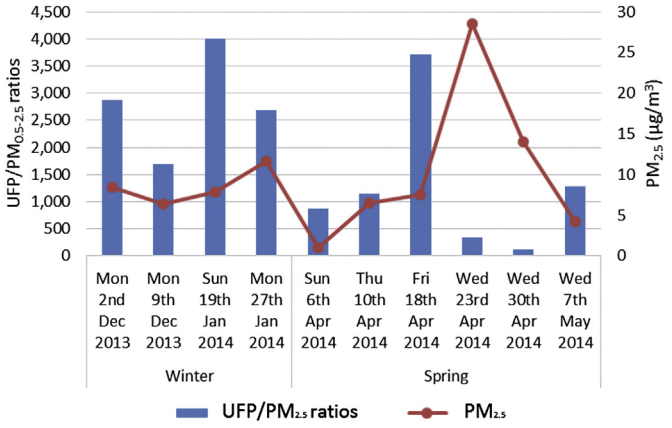

Fig. 4. Ratios between median UFP and $PM_{0.5\text{-}2.5}$ concentrations and average $PM_{2.5}$ concentration measured by TEOM-FDMS on each day. The ratios in the spring campaign were calculated from the static measurements. The same instruments were used to calculate the ratios in the winter campaign for the purpose of comparison between the ratios in the two campaigns.

The evidence presented here highlights that the effective management of $PM_{2.5}$ in the UK may require international scale cooperation in the control of emission sources. The good agreement between Dylos and TEOM-FDMS, together with the above analysis on the potential sources of high $PM_{2.5}$ episodes during each campaign, confirm that the Dylos is capable of measuring elevated $PM_{2.5}$ arising from both regional and local influences (Steinle et al., 2015). However, this analysis does not reveal the contribution from local traffic emission at street level. This is in line with the findings of Price et al. (2014) who showed that number concentration of particles greater than 262 nm was more closely related to meteorological conditions whilst UFP was more closely associated with traffic variables. Nevertheless this work presents a novel approach to apportioning $PM_{2.5}$ to local and regional sources by comparing the relationship between UFP and $PM_{2.5}$ with the help of back-trajectory analysis.

The IQR/median ratio was used to represent the variability of each pollutant in the spring measurements (Table 1b), in which the IQR/median ratio from the static measurements quantifies only the temporal variation between different times of a day, and the ratio in the mobile measurement reflects both spatial and temporal variation between different streets (with the former expected to be greater than the latter). Consistent with the observations in the winter measurements, the spatial variations of BC and UFP (average mobile IQR/median ratios of 1.05 and 1.08, respectively) were much larger than spatial variations of $PM_{0.5\text{-}2.5}$ (average mobile IQR/

median ratio of 0.29). The within-day variations of BC and UFP (range of static IQR/median ratios 0.14–2.92 and 0.26–1.08, respectively) were also generally larger than equivalent within-day variations of $PM_{0.5\text{-}2.5}$ (range of static IQR/median ratios 0.06–0.33) but were of varying magnitudes. This implies that changes in local traffic counts and atmospheric conditions within a day have more pronounced effects on variations in BC and UFP than on variations in $PM_{0.5\text{-}2.5}$. However $PM_{0.5\text{-}2.5}$ varied more between days (static IQR/median ratio for the whole spring campaign of 3.27) than BC and UFP (equivalent ratios of 1.77 and 2.19, respectively). Collectively this evidence suggests that the variability of all three PM metrics is subject to the varying background concentrations. For BC and UFP in particular, the geographical locations, namely proximity to trafficked roads, also contribute a significant part of the spatial variability. Contrary to another study (Sullivan and Pryor, 2014), which found that the spatial variability of $PM_{2.5}$ (defined as the relative standard deviation for the mobile measurements on different routes) was 2–3 times greater than the sub-daily temporal variability (defined as the RSD for the measurements when stationary), results from this study suggest that the spatial variability of $PM_{0.5\text{-}2.5}$ was of similar magnitude to the sub-daily temporal variability. Possible explanations of this discrepancy include the larger geographical area in the former study, and the potential for bias from a few extremely high concentrations when using RSD rather than IQR as an indicator for variability.

3.2. Pollutant concentration in relation to street topography and traffic counts

To understand the relationship between traffic counts and the pollutant concentration, reduced major axis (RMA) regression analyses were conducted between the mean of the concentrations on both sides of the road at each spot measurement in the winter campaign and traffic counts. Fig. 5 shows the scatter plots of BC and UFP against traffic counts grouped by the classification of each site. Correlations of 5-min average BC and UFP concentrations with traffic counts were moderate and highly significant ($r = 0.56$ and 0.39, respectively, $P < 0.01$, $n = 72$). $PM_{0.5\text{-}2.5}$ was not significantly correlated with traffic counts ($r = 0.17$, $n = 72$).

Ratios between mobile and static measurements in the spring campaign were calculated for each timestamp to represent the elevation in the pollutant concentration due to traffic. The distribution of the ratios for BC and UFP were grouped by streets and plotted in Fig. 6. Only ratios in the range 0–30 are shown in Fig. 6 to avoid extreme values skewing the plots. Median mobile/static ratios are higher on Sun 6th Apr, Thu 10th Apr and Wed 7th May 2014 than on other days, an observation more pronounced for BC than for UFP. These days coincide with the days with higher wind speed (Table S1), which explains the greater contrast between roadside and background concentrations as the high wind speed facilitates the mixing of clean air at the background site while the immediate roadside concentration still stays at relatively high level. A summary of the median values of the mobile/static measurement ratio for each street and the traffic density on each street are tabulated in Table 2. The data show a common pattern in the mobile/static ratios for BC and UFP: streets 2 and 3 had the highest median mobile/static ratios. Considering that the traffic density in streets 2 and 3 were not the highest among all the streets, the difference in street topography is the most likely explanation of the greater mobile/static ratios. Streets 2 and 3 are mostly characterised by street canyons with aspect ratios in the range of 0.6–1.3, whereas streets 1 and 4 are beside the Meadows urban park with an open terrain. As a result, dispersion in streets 2 and 3 is likely to be reduced compared to dispersion in streets 1 and 4. Similar results were reported in a study in Hong Kong characterised by high-rise buildings, where notably elevated BC and UFP concentrations were observed in deep street canyons compared to an open road although the traffic flow were significantly lower in street canyons (Rakowska et al., 2014).

The correlation of averaged BC and UFP concentration with traffic counts for each route segment was calculated in the spring campaign. BC was again significantly correlated with traffic counts ($r = 0.45$, $P < 0.01$, $n = 50$), but UFP was not ($r = 0.25$, $n = 48$, $P = 0.09$). The lower correlation in spring compared to winter campaign could be attributed to the fact that traffic counting on one side of the street in spring campaign may not have represented total traffic composition sufficiently. The low correlation between traffic count and UFP may have been caused by the secondary

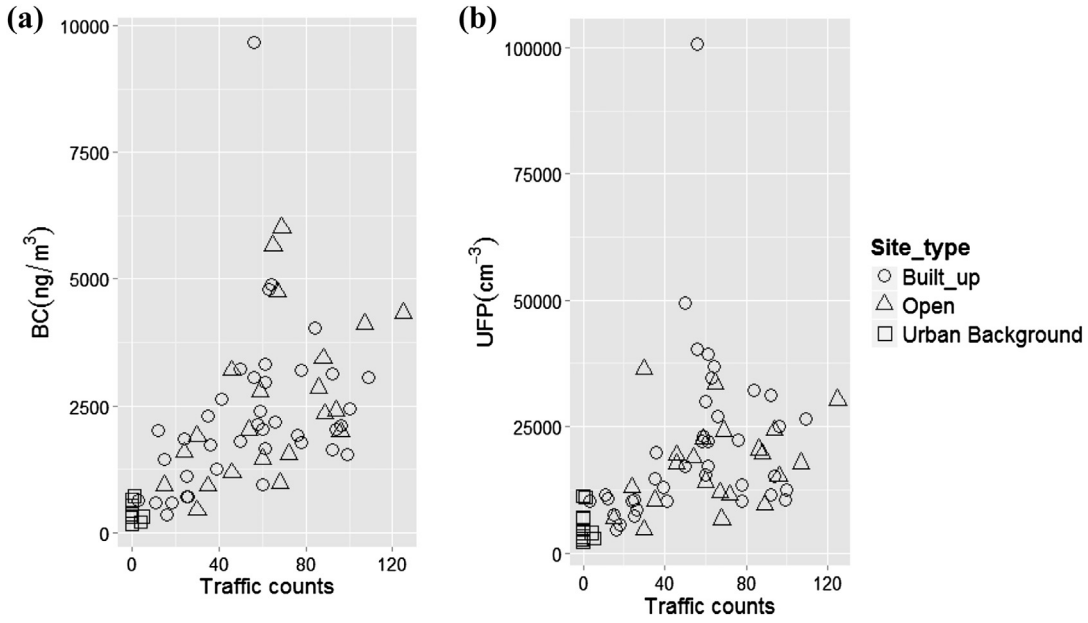

Fig. 5. Scatter plots of 5-min averaged (a) BC and (b) UFP concentrations vs. traffic counts at each measurement site in the winter campaign.

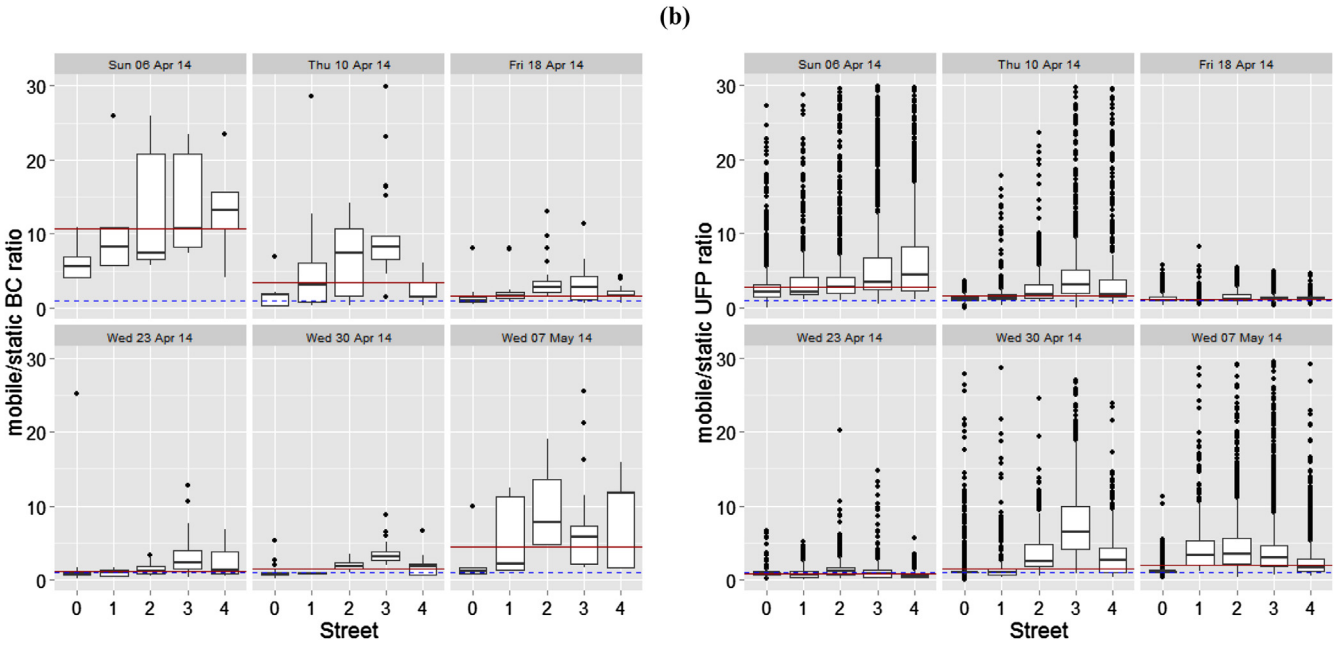

Fig. 6. Distributions of mobile/static measurement ratios in different streets for (a) BC, and (b) UFP, in the winter campaign. Solid red lines denote the median mobile/static measurement ratio for each day. The dashed blue line denotes a ratio of one to highlight the elevated concentrations on streets. Measurements at the static background were corrected based on the MA regression analyses results in Table S2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

particle formation mentioned in Section 3.1. This interpretation was supported by the increased correlation when the data from Friday was excluded ($r = 0.66$, $P < 0.01$, $n = 39$). Despite the moderate to high correlation coefficients of BC and UFP with traffic counts found in this study, mixed conclusions have been drawn in the literature for the relationships between traffic and BC or UFP as a result of characteristics of specific measurement sites, the consistency of traffic flow and the formation/transformation of particles governed by environmental conditions (Kumar et al., 2008; Peters et al., 2014; Price et al., 2014; Rakowska et al., 2014). Further investigation of relationships between BC, UFP and traffic would provide beneficial information to inform the potential use of nearby traffic flow data to predict BC and UFP concentrations. Street topography effects on BC and UFP concentrations are similarly important for further investigation.

3.3. Correlation between BC and UFP

The correlations between BC and UFP were evaluated using RMA regression. The results summarised in Table 3 were calculated from 1-min averaged concentrations with BC as the *y* variable and UFP as the *x* variable.

The correlations between BC and UFP during each week ranged between 0.49 and 0.77 and in all cases were significant ($P < 0.01$). During working and non-working days, slope coefficients were not significantly different between measurements taken on different sides of the road in the winter campaign or between mobile and static measurements in the spring campaign. The agreement of the BC/UFP relationship between roadsides or between busy roads and local background suggests that BC and UFP concentrations vary similarly as they disperse away from traffic sources. The slopes on non-working days were significantly lower than the slopes on working days, indicating a decrease in BC/UFP ratios on non-work days. This variation in the BC/UFP ratios was not only observed in the measurements taken near road but also in the static background measurements during the spring campaign. The reason for this variation in BC/UFP relationship is likely due to the decrease in heavy goods vehicles (HGV) on the road during the weekend as HGV had the lowest share on weekends (3.2%) compared with weekdays (~6.8%). This observation suggests that HGV contribute relatively more to BC than to UFP concentrations. Therefore a policy targeting on reduction of UFP may not be effective if it only focuses on restricting HGV. On the other hand policies aimed at reduction of BC should focus on controlling HGV emissions, which has direct implications for instance for the design and implementation of Low Emission Zones in urban areas. Considering that recent epidemiological studies suggested that BC was more strongly associated with harmful particle substances than was the $PM_{2.5}$ mass (Grahame et al., 2014; WHO, 2013), traffic control strategy targeting at the HGV may effectively contribute to

Table 2
Median ratios of mobile/static measurements in each street and the traffic density for each street.

Street	Median of the mobile/static ratios for all the days (range of medians for each day)		Mean traffic density (range of traffic density for each session) (traffic/km)
	BC	UFP	
0	1.1 (0.9–5.6)	1.1 (1.0–2.2)	0
1	1.4 (0.9–8.3)	1.4 (0.7–3.4)	27 (16–38)
2	3.0 (1.2–7.8)	2.0 (1.2–3.5)	33 (24–42)
3	4.2 (2.4–15.8)	2.8 (0.9–7.0)	27 (18–35)
4	1.8 (1.4–13.2)	1.8 (0.6–4.7)	39 (28–48)

Table 3
RMA regression analyses for 1-min averaged BC and UFP concentrations. The shading in the table represents the data collection period (grey: working days; white: non-working days). Left and right side of the road is defined with respect to the walking direction in the mobile measurements. ** indicates correlation at >99% significance.

r	Slope (ng × 10^{-6})	Intercept (ng/m^{-3})	Number of data points	r	Slope (ng × 10^{-6})	Intercept (ng/m^{-3})	Number of data points
Winter campaign							
Left side of road				Right side of road			
0.66**	0.15 (0.14–0.16)	−575 (−726 to −432)	833	0.65**	0.16 (0.15–0.17)	−737 (−900 to −583)	822
0.58**	0.08 (0.07–0.09)	149 (−1 to 283)	223	0.58**	0.11 (0.09–0.12)	137 (−43 to 298)	225
Spring campaign							
Mobile				Static			
0.49**	0.13 (0.12–0.14)	−102 (−289 to 69)	319	0.50**	0.11 (0.10–0.13)	148 (61–226)	270
0.62**	0.05 (0.04–0.06)	−48 (−158 to 50)	166	0.77**	0.04 (0.03–0.04)	−15 (−46 to 13)	181

alleviation of population health burdens arising from urban air pollution.

3.4. Study limitations

This paper describes design and interpretation of a pilot study to investigate potential drivers for the spatiotemporal variability of different particulate matter components in the urban environment. It is recognised that it may be limited in terms of giving exhaustive consideration and quantification of each individual driver on pollutant concentration, particularly the complex influence of local street topography. The absence of summer measurements could mean that the influence of photochemical particle formation may be underestimated (Ma and Birmili, 2015). Overall, however, this study has demonstrated that careful experimental design and data interpretation of short-term mobile measurements can identify the important drivers governing different metrics of PM concentration, and which also support findings in literature that use more sophisticated long term measurements.

4. Conclusions

Distributions of BC, UFP and $PM_{0.5\text{-}2.5}$ concentrations in the urban environment and the relationship between concentrations and potential influences (local traffic, street topography and synoptic meteorology) were studied through a combination of mobile and static measurements in the south of the city of Edinburgh, UK.

BC and UFP exhibited a high spatial variability in the urban environment, roughly 3 times greater than that of $PM_{0.5\text{-}2.5}$; however $PM_{0.5\text{-}2.5}$ had the highest inter-daily variability. Very high BC and UFP concentrations were frequently measured at roadside and sometimes exceeded 3 times the street median concentration. However the median BC and UFP concentrations measured on the streets were not greatly influenced by the frequent spikes and were on average approximately double the median measured on the footpath. $PM_{0.5\text{-}2.5}$ did not show a consistent elevated concentration in the streets in comparison with the footpath. Both geographical locations and varying background concentrations had important effects on the BC and UFP concentrations. Variation in $PM_{0.5\text{-}2.5}$ concentrations was largely influenced by regional sources, although local sources also contributed to a lesser extent.

Both BC and UFP were highly correlated with traffic counts. $PM_{0.5\text{-}2.5}$ showed very low correlation with the traffic counts. Significant difference was found between the slopes of BC vs. UFP regression analyses between working days and non-working days. During non-working days the reduction of HGV flows results in a decrease in the BC/UFP ratio, suggesting that HGV may contribute more to BC than to UFP concentrations. Therefore control of HGVs may be effective in reducing the negative health effects associated with BC. $PM_{2.5}$ was observed in a few instances to have a large background component, the variability of which may not be well accounted for by only using local sources as indicators. Therefore international cooperation in control of emission sources is important for $PM_{2.5}$ management in the UK. Furthermore, this study indicates that the proximity to the road as a primary indicator for the assessment of exposure to air pollutants is highly dependent on the pollutant being considered.

Acknowledgement

Hao Wu acknowledges funding from the University of Edinburgh and the NERC Centre for Ecology & Hydrology (NERC CEH project number NEC04544). The CPC and AE51 instruments were purchased under a NERC multi-institution grant (NE/1007822/1). We thank Hugo Man for assistance in conducting the field measurements and Christopher Malley for help in calculating back trajectories. We thank Defra for data from the UK AURN.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.04.059.

References

AQEG, 2012. Fine Particulate Matter ($PM_{2.5}$) in the United Kingdom. Air Quality Expert Group, UK Department for Environment. Food and Rural Affairs, London. PB13837. http://uk-air.defra.gov.uk/library/reports?report_id=727.
Ayers, G.P., 2001. Comment on regression analysis of air quality data. Atmos. Environ. 35, 2423–2425.
Carslaw, D.C., Ropkins, K., 2012. openair — An R package for air quality data analysis. Environ. Model. Softw. 27–28, 52–61.
Dft, 2015. City of Edinburgh [WWW Document]. Dep. Transp. – Traffic Counts. http://www.dft.gov.uk/traffic-counts/cp.php?la=City+of+Edinburgh.
Grahame, T.J., Klemm, R., Schlesinger, R.B., 2014. Public health and components of particulate matter: the changing assessment of black carbon. J. Air Waste Manag. Assoc. 64, 620–660.
Hagler, G.S.W., Yelverton, T.L.B., Vedantham, R., Hansen, A.D.A., Turner, J.R., 2011. Post-processing method to reduce noise while preserving high time resolution in aethalometer real-time black carbon data. Aerosol Air Qual. Res. 11, 539–546.
Heal, M.R., Kumar, P., Harrison, R.M., 2012. Particles, air quality, policy and health. Chem. Soc. Rev. 41, 6606.
HEI, 2010. Traffic-related Air Pollution: a Critical Review of the Literature on Emissions, Exposure, and Health Effects. Health Effects Institute.
Hoek, G., Beelen, R., de Hoogh, K., Vienneau, D., Gulliver, J., Fischer, P., Briggs, D., 2008. A review of land-use regression models to assess spatial variation of outdoor air pollution. Atmos. Environ. 42, 7561–7578.
Hoek, G., Meliefste, K., Cyrys, J., Lewné, M., Bellander, T., Brauer, M., Fischer, P., Gehring, U., Heinrich, J., van Vliet, P., Brunekreef, B., 2002. Spatial variability of fine particle concentrations in three European areas. Atmos. Environ. 36, 4077–4088.
Kassomenos, P.A., Vardoulakis, S., Chaloulakou, A., Paschalidou, A.K., Grivas, G., Borge, R., Lumbreras, J., 2014. Study of PM10 and PM2.5 levels in three European cities: analysis of intra and inter urban variations. Atmos. Environ. 87, 153–163.
Knibbs, L.D., Cole-Hunter, T., Morawska, L., 2011. A review of commuter exposure to ultrafine particles and its health effects. Atmos. Environ. 45, 2611–2622.

Kumar, P., Fennell, P., Britter, R., 2008. Measurements of particles in the 5-1000 nm range close to road level in an urban street canyon. Sci. Total Environ. 390, 437–447.

Ma, N., Birmili, W., 2015. Estimating the contribution of photochemical particle formation to ultrafine particle number averages in an urban atmosphere. Sci. Total Environ. 512–513, 154–166.

Patton, A.P., Perkins, J., Zamore, W., Levy, J.I., Brugge, D., Durant, J.L., 2014. Spatial and temporal differences in traffic-related air pollution in three urban neighborhoods near an interstate highway. Atmos. Environ. 99, 309–321.

Peters, J., Van den Bossche, J., Reggente, M., Van Poppel, M., De Baets, B., Theunis, J., 2014. Cyclist exposure to UFP and BC on urban routes in Antwerp. Belg. Atmos. Environ. 92, 31–43.

Pinto, J.P., Lefohn, A.S., Shadwick, D.S., 2004. Spatial variability of PM2.5 in urban areas in the United States. J. Air Waste Manag. Assoc. 1995 (54), 440–449.

Price, H.D., Arthur, R., BéruBé, K.A., Jones, T.P., 2014. Linking particle number concentration (PNC), meteorology and traffic variables in a UK street canyon. Atmos. Res. 147–148, 133–144.

Rakowska, A., Wong, K.C., Townsend, T., Chan, K.L., Westerdahl, D., Ng, S., Močnik, G., Drinovec, L., Ning, Z., 2014. Impact of traffic volume and composition on the air quality and pedestrian exposure in urban street canyon. Atmos. Environ. 98, 260–270.

R Core Team, 2014. R: a Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria. http://www.R-project.org/.

Reche, C., Querol, X., Alastuey, A., Viana, M., Pey, J., Moreno, T., Rodríguez, S., González, Y., Fernández-Camacho, R., de la Campa, A.M.S., de la Rosa, J., Dall'Osto, M., Prévôt, A.S.H., Hueglin, C., Harrison, R.M., Quincey, P., 2011. New considerations for PM, black Carbon and particle number concentration for air quality monitoring across different European cities. Atmos. Chem. Phys. 11, 6207–6227.

Ruths, M., von Bismarck-Osten, C., Weber, S., 2014. Measuring and modelling the local-scale spatio-temporal variation of urban particle number size distributions and black carbon. Atmos. Environ. 96, 37–49.

Sandradewi, J., Prévôt, A.S.H., Szidat, S., Perron, N., Alfarra, M.R., Lanz, V.A., Weingartner, E., Baltensperger, U., 2008. Using aerosol light absorption measurements for the quantitative determination of wood burning and traffic emission contributions to particulate matter. Environ. Sci. Technol. 42, 3316–3323.

Skyllakou, K., Murphy, B.N., Megaritis, A.G., Fountoukis, C., Pandis, S.N., 2014. Contributions of local and regional sources to fine PM in the megacity of Paris. Atmos. Chem. Phys. 14, 2343–2352.

Steinle, S., Reis, S., Sabel, C.E., 2013. Quantifying human exposure to air pollution – moving from static monitoring to spatio-temporally resolved personal exposure assessment. Sci. Total Environ. 184.

Steinle, S., Reis, S., Sabel, C.E., Semple, S., Twigg, M.M., Braban, C.F., Leeson, S.R., Heal, M.R., Harrison, D., Lin, C., Wu, H., 2015. Personal exposure monitoring of PM2.5 in indoor and outdoor microenvironments. Sci. Total Environ. 508, 383–394.

Sullivan, R.C., Pryor, S.C., 2014. Quantifying spatiotemporal variability of fine particles in an urban environment using combined fixed and mobile measurements. Atmos. Environ. 89, 664–671.

Vieno, M., Heal, M.R., Hallsworth, S., Famulari, D., Doherty, R.M., Dore, A.J., Tang, Y.S., Braban, C.F., Leaver, D., Sutton, M.A., Reis, S., 2014. The role of long-range transport and domestic emissions in determining atmospheric secondary inorganic particle concentrations across the UK. Atmos. Chem. Phys. 14, 8435–8447.

Warton, D.I., Wright, I.J., Falster, D.S., Westoby, M., 2006. Bivariate line-fitting methods for allometry. Biol. Rev. 81, 259–291.

WHO, 2013. Review of Evidence on Health Aspects of Air Pollution – REVIHAAP Project: Technical Report. World Health Organisation, Copenhagen, Denmark. http://www.euro.who.int/__data/assets/pdf_file/0004/193108/REVIHAAP-Final-technical-report-final-version.pdf.