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Estimation of spatial patterns of urban air pollution over a 4-week period from repeated 5-min measurements

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Short communication

Estimation of spatial patterns of urban air pollution over a 4-week period from repeated 5-min measurements

ATMOSPHERIC
ENVIRONMENT

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HIGHLIGHTS

- Peripatetic BC and PN measurements made weekly for 4 weeks at 18 urban sites.
- 5-min BC measurements were highly correlated with weekly $NO₂$.
- Average of 5 repeated 5-min BC measurements explained 75% variation of $NO₂$.
- Short duration BC provided information on longer-term $NO₂$ spatial contrasts.

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GRAPHICAL ABSTRACT highlights grap hical abstract

ABSTRACT abstract

Determination of intra-urban spatial variations in air pollutant concentrations for exposure assessment requires substantial time and monitoring equipment. The objective of this study was to establish if shortduration measurements of air pollutants can be used to estimate longer-term pollutant concentrations. We compared 5-min measurements of black carbon (BC) and particle number (PN) concentrations made once per week on 5 occasions, with 4 consecutive 1-week average nitrogen dioxide $(NO₂)$ concentrations at 18 locations at a range of distances from busy roads in Glasgow, UK. 5-min BC and PN measurements (averaged over the two 5-min periods at the start and end of a week) explained $40-80\%$, and $7-64\%$ respectively, of spatial variation in the intervening 1-week $NO₂$ concentrations for individual weeks. Adjustment for variations in background concentrations increased the percentage of explained variation in the bivariate relationship between the full set of $NO₂$ and BC measurements over the 4-week period from 28% to 50% prior to averaging of repeat measurements. The averages of five 5-min BC and PN measurements made over 5 weeks explained 75% and 33% respectively of the variation in average 1 week NO₂ concentrations over the same period. The relatively high explained variation observed between BC and NO₂ measured on different time scales suggests that, with appropriate steps to correct or average out temporal variations, repeated short-term measurements can be used to provide useful information on longer-term spatial patterns for these traffic-related pollutants.

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1. Introduction

A major challenge in quantifying the effect of air pollution on human health is the resource required reliably to measure spatial

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Abbreviation: BC, Black carbon; PN, Particle Number; AADF, annual average daily flow; PDT, passive diffusion tube; NO₂, Nitrogen Dioxide; RMA, reduced major axis.

and temporal variations in pollutant concentrations within urban environments ([Hoek et al., 2008\)](#page-7-0). The development of lightweight, lower-power portable monitoring equipment provides new opportunities to design monitoring studies that supplement static monitoring networks by using mobile measurements. Two approaches are possible: continuously mobile monitoring, where the monitoring equipment is moved throughout the duration of the study; and peripatetic monitoring, where mobile equipment is deployed at specific sites for short time periods before moving to another site.

Peripatetic measurements allow collection of observations through a monitoring network over a period of time and over relatively large areas with limited equipment. This approach has been used to monitor air pollution at sequential locations in studies in Canada ([Abernethy et al., 2013; Deville Cavellin et al., 2016;](#page-7-0) [Larson et al., 2009\)](#page-7-0), Germany ([Merbitz et al., 2012\)](#page-8-0), India ([Saraswat et al., 2013\)](#page-8-0), the Netherlands ([Klompmaker et al., 2015\)](#page-7-0), Spain ([Rivera et al., 2012\)](#page-8-0), Switzerland [\(Ragettli et al., 2014\)](#page-8-0) and the USA [\(Riley et al., 2016\)](#page-8-0). A limitation with this approach is the difficulty in accounting for fluctuating background concentrations, although this can be mitigated by using a static background site during the study ([Hoek et al., 2008; Klompmaker et al., 2015](#page-7-0)).

The objective of our study was to examine the quantitative relationships between short-term peripatetic measurements made with handheld equipment and longer-term average spatial air pollutant patterns, to assess if one can be used as a surrogate for the other. We combined peripatetic 5-min 'spot' measurements of black carbon (BC) and particle number (PN) [using portable lowpower equipment] with weekly nitrogen dioxide $(NO₂)$ measurement [using passive diffusion tubes (PDT)] over four 1-week periods at 18 sites of varying distance from major roads in Glasgow, UK. Five-minute static measurements were made at each site during deployment and retrieval of the PDTs. The combination of PDT and peripatetic measurements enabled investigation of spatial correlations between different pollutants at different averaging periods.

2. Methods

2.1. Monitoring plan

The study was conducted in the city of Glasgow (population ~ 600,000) in the west of Scotland (55.87 \textdegree N, 4.26 \textdegree W), for four consecutive weeks beginning on 24 October 2013. Eighteen monitoring sites were selected in a mixed residential and commercial area in the West End of the city to provide a range of local traffic influence (Fig. 1). The two busiest roads in the study area, Byres Road and Dumbarton Road, have annual average daily flows (AADF) of approximately 10,000 vehicles day⁻¹. A background site (Site 18) in a nearby park provided measurements free from immediate influence of local traffic sources.

Duplicate $NO₂$ PDTs were located at each site and changed approximately weekly at times that avoided adverse weather to avoid damage to real-time equipment. Therefore, weeks 1 and 2 spanned 8 and 6 days respectively, while weeks 3 and 4 spanned 7 days. During PDT exchange, while stationary at each site, 5-min peripatetic measurements were made using handheld BC and PN instruments (Section [2.2\)](#page-3-0). PDT changeovers began around 08:00 local time and took approximately 2.5 h to complete. All real-time instrument clocks were synchronized prior to measurements. Peripatetic measurements were made during this time of the morning when many people were traveling on roads to get to work to maximise the range of observed concentrations. To reduce the possibility of systematic bias, sites 1 to 17 were visited in opposite order on alternate weeks (starting with site 1 in w/c week 1). Because of its distance from the other sites, the background site, 18, was always visited last. A duplicate PN instrument at the background site provided an indication of changes in background

Fig. 1. Locations in the West End of Glasgow of NO₂ passive diffusion tubes and 'spot' monitoring locations for BC and PN concentrations.

concentration during each measurement period (Supplementary information $-$ Fig. S4). A duplicate BC instrument was not available, so for consistency we made background adjustments for both pollutants using 5-min measurements made at site 18 at the end of each measurement period.

In this Short communication the notation 'week X' and abbreviation 'wX' refer to PDT measurements throughout week X or to 'weekly spot' measurements derived from averaging the mobile measurements made at the beginning and end of week X, while 'w/ c week X' and abbreviation 'w/cX' refer to mobile measurements made at the start of week X only. For example, 'w/c week 5' refers to mobile measurements made at the end of the fourth week of the study when PDTs were collected for the final time.

2.2. Instruments and data processing

BC concentrations were measured using a microaethalometer (Model AE51, Aethlabs, San Francisco, CA) carried in a backpack, with the manufacturer-supplied 1 m conductive plastic tubing inlet mounted on the shoulder strap. BC was recorded at 1-min resolution during w/c week 1, and 1-s resolution during subsequent weeks. This change made it easier to synchronise arrival and departure times at PDT sites with logged data. At 1-s temporal resolution microaethelometers are prone to measurement artifacts ([Hagler, 2011](#page-7-0)). Consequently 1-s BC data were processed using an optimised noise adjustment (ONA) method (Δ ATN = 0.01) to retain the highest possible temporal resolution ([AethLabs, 2013; Hagler,](#page-7-0) [2011](#page-7-0)). A second adjustment, to account for non-linear response with increasing BC deposition, was also applied ([Apte et al., 2011\)](#page-7-0):

$$
BC = BC_0 \left(0.88 \exp \left(-\frac{ATN}{100} \right) + 0.12 \right)^{-1}
$$

where $BC =$ adjusted BC concentration, $BC₀ =$ unadjusted BC concentration, and $ATN =$ attenuation value from the instrument. A single filter strip was sufficient for all measurements and gave an ATN value of <50 at the conclusion of the study. The AE51 instrument was evaluated by deployment next to an AE22 aethalometer used for black carbon measurements at the UK government Automatic Urban and Rural Network (AURN) monitoring site at Townhead, Glasgow (Fig. S2).

PN was measured using two handheld condensation particle counters (CPC 3007, TSI Inc., Shoreview, MN). Before each set of measurements the CPCs were checked for zero reading, supplied with fresh isopropyl alcohol, and allowed to warm up for 10 min. Precision of the duplicate PN instruments was assessed by walking them together through urban environments with a similar range of pollutant concentrations to those in this study. Duplicate instruments exhibited a high degree of precision ($R^2 = 0.93$, Fig. S1) and <2% normalised mean bias between the paired instruments.

Palmes NO₂ PDTs were deployed in duplicate at 2.5 m elevation at each site. PDTs were prepared as a single batch at the beginning of the campaign by dipping stainless steel mesh grids into 50% triethanolamine-acetone solution [\(Heal, 2008](#page-7-0)) and stored double bagged in a refrigerator pre and post deployment. Two 'travel' blanks were carried during deployment and retrieval of PDTs, and kept in a laboratory refrigerator during the intervening period. Two 'field' blanks were deployed close to site 13, and two 'laboratory' blanks were kept in the refrigerator during the exposure period. PDTs and blanks were analysed within 3 or 4 days of retrieval using a standard protocol ([Targa et al., 2008\)](#page-8-0). Laboratory and travel blanks showed no significant concentration values. The mean relative standard deviation $(\pm 1 \text{ sd})$ for all 70 duplicate PDT measurements was 6.4 (\pm 6.8)%, comparable with that reported in the literature (e.g. [Lewne et al., 2004](#page-8-0)). Four out of 144 (3%) PDTs were lost during measurements, consisting of pairs of duplicate tubes lost in week 1 and week 4 from sites 17 and 16 respectively. Consequently statistics in weeks 1 and 4 were not fully comparable with other weeks, as data from two of the highest concentration sites were missing during these weeks.

We used four approaches to assess if short-term measurements could provide useful information on longer-term spatial trends in pollutant concentrations. Firstly, we compared 5-min BC and PN 'spot' measurements made in each week to the average of all 5-min measurements for BC and PN over the study. Secondly, we calculated the average of BC and PN spot measurements made at the start and end of each week (subsequently referred to as 'weekly spot' measurements) and compared this average to weekly $NO₂$ concentrations measured by PDTs throughout the intervening period. Thirdly, we corrected weekly 'spot' BC and weekly $NO₂$ concentrations to allow for changes in background concentrations measured at site 18 at the end of each measurement period. This was done by using a 'difference' method [\(Klompmaker et al., 2015\)](#page-7-0) that involved: (a) computation of the overall mean concentration for the full set of measurements at the background site (site 18) for each pollutant ($C_{ref,ave}$); (b) computation of differences between period specific measurements at the background site ($C_{ref,t}$) and the estimated overall background mean $(C_{ref,ave})$ for each pollutant for each period (t) $(C_{diff,ref,t} = C_{ref,ave} - C_{ref,t})$; (c) correction of the period measurement at each site (x) by addition of the difference calculated in step (b) ($C_{x,t,corrected} = C_{x,t,measured} + C_{diff,ref,t}$). A 'ratio' method of temporal adjustment [\(Klompmaker et al., 2015](#page-7-0)) was also examined but found to produce less consistent reduction in withinsite/between-site variance ratios [\(Table 2](#page-4-0)); therefore most of our analyses with temporal adjustments were focused on the difference method. In a fourth approach we examined the bivariate relationships between estimates of the overall averages of $NO₂$, BC and PN concentrations for the 4-week period across the 18 sites. Reduced major axis (RMA) regression was used to compare pollutant metrics in the above approaches [\(Ayers, 2001](#page-7-0)). One-way analysis of variance (ANOVA) was used to compare within-site (temporal)/between-site (spatial) variance ratios ([Klompmaker](#page-7-0) [et al., 2015](#page-7-0)).

3. Results and discussion

3.1. NO₂, BC and PN by site

Descriptive statistics and discussion of the time series of measurements are provided in the Supplementary Information.

Relatively high $NO₂$ concentrations were consistently observed across all weeks at sites closest to main roads (sites 1, 6, 7, 16, 17) ([Figs. 1 and 2](#page-2-0)). The lowest $NO₂$ concentration each week was observed at the background site (Site 18), where $NO₂$ concentration varied markedly between weeks, but was always $2-3$ times lower than the maximum observed concentration for each week [\(Fig. 2,](#page-5-0) Table $S1$). Despite large variations in average $NO₂$ concentrations between weeks, spatial patterns of relative concentrations across the sites remained consistent from week to week (concentrations were highly correlated between pairs of successive weeks $(R^2 = 82\%, 88\% \text{ and } 82\%)$.

5-min averaged 'spot' measurements for BC and PN demonstrated qualitatively similar spatial patterns to $NO₂$ [\(Fig. 2\)](#page-5-0). However, the spot measurements showed a less consistent spatial pattern between successive weekly measurements than was observed for NO₂ (R^2 ranges of 29–81% and 0–29% for BC and PN respectively).

Background concentrations measured at site 18 at the end of each weekly monitoring period were, on average across all weeks,

Table 1

Reduced major axis regression statistics between longer-term average pollution measurements and 5-min BC (μ g m $^{-3}$) and 5-min PN (#x 10 3 cm $^{-3}$) spot measurements.

^a NO₂ measurements at background site are listed alongside intercepts for non-background-adjusted NO₂ vs. BC regression lines.
^b 'ave' represents the average of all NO₂ PDT and BC/PN spot measurements over the f ference' method described in Methods Section [2.2](#page-3-0) (based on method described by [Klompmaker et al., 2015](#page-7-0)).

Table 2

Comparison of within:between site variance ratios from this study, and other European studies (MUSiC, ESCAPE, RUPIOH, VE³SPA) summarised by [Klompmaker et al. \(2015\)](#page-7-0).

Study:	Repeats \times duration	NO ₂	NO ₂ (adi ^a)	BC	BC(adi ^a)	PN	PN(adi ^a)
Present study ('ratio' method ^a)	5×5 -min BC & PN 4×1 -week NO ₂	0.14	0.31	0.28	0.13	1.17	0.64
Present study ('difference' methoda')	5×5 -min BC & PN 4×1 -week NO ₂	0.14	0.05	0.28	0.21	1.17	0.77
MUSiC	3×30 - min PN			3.25	2.44	2.21	2.17
ESCAPE	3×14 -day PM _{2.5} absorbance			0.39	0.09		
RUPIOH	3×1 -day PN					0.5	0.31
VE ³ SPA	6×4 -day PM _{2.5} absorbance			2.55	0.69		

 $^{\text{a}}$ Adj = adjustment for temporal variation. For overview of different methods used to adjust for temporal variation see [Klompmaker et al. \(2015\)](#page-7-0).

~65%, 40% and 30% of the mean concentration of all other sites for NO2, PN and BC respectively suggesting that, of those pollutants measured in this study, BC was the metric with spatial variations that are most influenced by proximity to local traffic sources.

3.2. Longer-term predictions from 5 min measurements

The linear relationship between 5-min PN measurements and the average of five 5-min spot PN was not significant for 1 out of 5 weeks (Table 1a). Relatively low week-to-week correlations between PN measurements (Section [3.1](#page-3-0)) may have resulted from changes in atmospheric processes that determine the formation of ultrafine particles through changes in meteorology between and within weeks. Meteorological conditions also influence $NO₂$ concentrations but the influence would have been reduced for the 1 week averaged PDT measurements compared with the shortterm PN measurements. Correlations between 'weekly spot' PN and 1-week $NO₂$ concentrations were not significant on 2 of 4 weeks, and explained $<$ 25% of variation in 1-week NO₂ concentrations during all but one week (Table 1b). Consequently the remainder of this Short communication focuses on the more clearly observed relationships between BC and NO2.

Fig. 2. Concentrations of (A) NO₂, (B) BC and (C) PN at each site. NO₂ concentrations are 1-week averages and BC and PN concentration are averages of 5-min 'spot' measurements.

5-min BC measurements were significantly associated with the average of five 5-min spot measurements taken once per week (average explained variation 55%, range $44-87%$) [\(Table 1](#page-4-0)a). 'Weekly spot' measurements of BC explained between 40% and 80% (average $= 62\%)$ of the variation in 1-week NO₂ concentrations ([Table 1b](#page-4-0), [Fig. 3](#page-6-0)a). The lowest explained variation was observed during week 4 and may have resulted from limited variation in BC concentration and missing data at one of the higher concentration NO2 sites (Site 16). The regression slope and intercept varied between weeks, with the y-axis intercept providing a good approximation of the background $NO₂$ concentration measured at Site 18 ([Table 1\)](#page-4-0). This suggests that (subject to confirmation using observations for larger areas and longer periods) it may be possible to estimate urban background $NO₂$ concentration using short-term BC measurements alongside weekly NO₂ PDTs.

Correlation between background-adjusted BC and $NO₂$ was highly significant and explained 50% of the variation in weekly $NO₂$ concentrations for the full set of measurements $(cf. 28\%$ explained variation prior to background adjustment) ([Table 1,](#page-4-0) [Fig. 3\)](#page-6-0). The overall average of 5-min BC spot measurements over the full study period (5-min measurements repeated 5 times over 4 weeks) explained 75% of the variation in overall average NO₂

Fig. 3. RMA linear regression analyses between NO₂ and BC metrics: (A) Unadjusted 1-week NO₂ vs. 1-week (start & end) BC; (B) 1-week NO₂ adjusted by difference method vs. 1-week (start and end) BC adjusted by difference method [\(Klompmaker et al., 2015\)](#page-7-0); (C) 4-week average NO₂ vs. average of BC w/c 1, 2, 3, 4, 5; (D) 4-week average NO₂ vs. average of BC w/c 1, 3, 5; (E) 4-week average $NO₂$ vs. average of BC w/c 1, 5.

concentrations [\(Table 1,](#page-4-0) Fig. 3c). Averaging selected subsets of repeated BC spot measurements interspersed evenly within the 5 measurement periods (to simulate a lower repeat peripatetic measurement frequency) resulted in a lower percentage of explained variation in overall average $NO₂$ concentrations (69% and 59% using weeks 1, 3 $\&$ 5 and weeks 1 $\&$ 5 respectively (Fig. 3d and e)).

Our results and conclusions are broadly coherent with comparisons of mobile real-time and static passive measurements of traffic-related pollutants in Baltimore, USA using different measurement approaches over different time and geographical scales ([Riley et al., 2016\)](#page-8-0). Our findings can also be set in the context of quantitative analyses of within-site/between-site variance ratios for BC and PN peripatetic measurements in the Netherlands ([Klompmaker et al., 2015](#page-7-0)). Our 5-min peripatetic observations have lower within-site/between-site variance ratios (i.e. exhibit more temporal consistency in spatial patterns) than 30-min peripatetic observations in the MUSiC study in the Netherlands ([Table 2\)](#page-4-0). The magnitudes of the ratios we observed are relatively close to those observed with 14 day measurement periods in the European ESCAPE project ([Table 2\)](#page-4-0). The reasons for the relatively limited temporal variation in the observations in our study are not fully clear, but may be related to the relatively small geographical area and short time period over which measurements were conducted. The relatively limited temporal variation in our measurements are also consistent with temporally persistent spatial variations in $NO₂$

and $O₃$ concentrations observed in PDT measurements in the nearby city of Edinburgh, UK [\(Lin et al., 2016](#page-8-0)).

Collectively the relatively high correlations observed between NO2 and BC measurements, and relatively low within-site/ between-site variance ratios suggest that short-term measurements with limited repetition are capable of partly characterising pollution concentration gradients in the urban environment. However, some limitations are relevant for consideration. Firstly, in the absence of continuous longer-term measurements of BC to compare with short-duration measurements, our study made use of the relationship between BC and $NO₂$ to assess the effectiveness of 5-min measurements for estimation of longer-term spatial contrasts. Other studies have shown BC and $NO₂$ to be highly correlated over extended time periods (Durant et al., 2014). Secondly, each week of mobile measurements was completed in approximately 2.5 h, around the time of the morning rush hour. Our measurements may have been affected by changing traffic and meteorological conditions. We attempted to minimise systematic bias by reversing the order in which sites were visited on alternate weeks. Thirdly, there is uncertainty regarding the optimal duration for 'spot' measurements, and whether the sampling period should be the same for all site classifications. We observed limited variation in concentrations during the 5-min 'spot' measurements at sites adjacent to roads with lower traffic flows (Fig. S3), where a shorter duration 'spot' measurement may have been sufficient. Conversely, 'spot' measurements made at sites near higher and variable traffic flows were more variable (Fig. S3) and may benefit from a longer measurement period. Additionally, when measurements are made in areas where traffic is influenced by local traffic signals, it may be appropriate to increase the 'spot' measurement period to encompass the full cycle of the traffic signals.

4. Conclusions

This study compared 5-min 'spot' measurements of black carbon (BC) and particle number (PN) concentrations, measured at weekly intervals at 18 locations in the city of Glasgow, against 1 week measurements of $NO₂$ concentrations. On average, 5-min BC measurements during individual measurement periods explained 55% variation of the overall average of five 5-min spot measurements taken once per week over the 4-week period. BC measurements of 5-min duration at the beginning and end of weeks explained 40–80% of spatial variations in $NO₂$ during the intervening 1-week periods. Equivalent measurements of PN explained 7-64% of 1-week $NO₂$ spatial variations. After adjusting for changes in background $NO₂$ and BC concentrations, spot measurements of BC and PN conducted repeatedly over a 4-week period, explained 50% and 24% respectively of the spatial variation in the complete set of corresponding 1-week $NO₂$ concentrations. The average of 5 replicate 5-min BC and PN spot measurements explained 75% and 33% respectively of the spatial variation in 4-week average $NO₂$ concentrations. Reducing the number of replicate peripatetic BC measurements from 5 to 3 and 2 replicates reduced the percentage of explained variation in spatial variation in 4-week average $NO₂$ concentrations to 67% and 59% respectively. Collectively these observations (with appropriate allowance for their relatively limited duration and spatial extent) suggest that short-term peripatetic measurements can be used to estimate longer term spatial contrasts in traffic-related air pollution provided that appropriate steps are taken to correct or average out temporal variations.

Author contributions

Iain Beverland, Jonathan Gillespie and Nicola Masey contributed

to the design of the study. Jonathan Gillespie, Nicola Masey conducted data collection. The first draft of the manuscript was manuscript was written by Jonathan Gillespie and all other authors contributed to discussions on data analysis and revisions of the paper. All authors have given approval to the final version of the manuscript.

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Appendix A. Supplementary Information

Supplementary information related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2016.11.035>.

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