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Quantifying spatiotemporal variability of fine particles in an urban environment using combined fixed and mobile measurements

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HIGHLIGHTS

- $PM_{2.5}$ measured in Indianapolis indicates a strong signature from mobile emissions.
- Intra-urban $PM_{2.5}$ variability is the same magnitude as between urban-rural differences.
- Spatial variability is greater than temporal variability on short time scales.
- ~1/3 of hours with high concentrations at one site were not observed at other sites.

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ABSTRACT

Spatiotemporal variability of fine particle concentrations in Indianapolis, Indiana is quantified using a combination of high temporal resolution measurements at four fixed sites and mobile measurements with instruments attached to bicycles during transects of the city. Average urban $PM_{2.5}$ concentrations are an average of ~3.9–5.1 μg m^{-3} above the regional background. The influence of atmospheric conditions on ambient $PM_{2.5}$ concentrations is evident with the greatest temporal variability occurring at periods of one day and 5–10 days corresponding to diurnal and synoptic meteorological processes, and lower mean wind speeds are associated with episodes of high $PM_{2.5}$ concentrations. An anthropogenic signal is also evident. Higher $PM_{2.5}$ concentrations coincide with morning rush hour, the frequencies of $PM_{2.5}$ variability co-occur with those for carbon monoxide, and higher extreme concentrations were observed mid-week compared to weekends. On shorter time scales (<one day), spatial variability is 2–3 times greater than temporal variability. Spatial variability along the transect (within 10 km of the fixed measurement sites) was within ±3.6 μg m^{-3} of the stationary measurements (once the temporal variability is removed). Localized extreme values of $PM_{2.5}$ concentrations ranged in the spatial dimension from a few hundred meters up to 2 km, and contributed an average of 4.4 μg m^{-3} to ambient concentrations.

1. Introduction

Human exposure to fine particles ($PM_{2.5}$, i.e. particles with diameters ≤ 2.5 μm) is associated with a range of negative health outcomes (Straif et al., 2013). Regulatory standards have been implemented to limit annual average and daily averaged $PM_{2.5}$ concentrations to reduce the excess mortality, and non-fatal cardiovascular and respiratory problems associated with chronic exposure to elevated $PM_{2.5}$ concentrations (Dockery et al., 1993; Pope III et al., 2002). Acute exposure to elevated $PM_{2.5}$ concentrations on time scales of minutes to hours has also been linked to fatal and non-fatal cardiovascular episodes (Brook et al., 2010; Peters et al., 2001), and although development of a 1-h standard has been proposed (Michaels and Kleinman, 2000), no regulatory standard is currently in place to reduce this public health hazard.

Air quality degradation due to elevated $PM_{2.5}$ concentrations is widespread. For example, over 90% of the urban population in the European Union is estimated to be exposed to $PM_{2.5}$ concentrations above World Health Organization guidelines (European Environment Agency, 2013). The research presented herein is focused on the city of Indianapolis in the Midwestern USA. In 2008 over 26 million Midwestern residents inhabited counties that failed the National Ambient Air Quality Standard (NAAQS) for $PM_{2.5}$

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(Pryor, 2013). Marion County (where Indianapolis is located) and four adjacent counties failed the annual NAAQS of 15 μg m^{-3} from 2005 to 2012 (Fig. 1a) (data from http://www.epa.gov/airquality/greenbk/), and may be in nonattainment for the new standard of 12 μg m^{-3} promulgated in 2013 (Esworthy, 2013).

Ambient $PM_{2.5}$ concentrations are a function of prevailing meteorological conditions and primary particle and precursor gas emissions. Most research has indicated chronic exposure to elevated $PM_{2.5}$ concentrations occurs primarily in urban environments, leading to research designed to quantify spatiotemporal variability of concentrations and public exposure to particulate matter on urban and intra-urban scales (Hoek et al., 2002; Kim et al., 2005; Pinto et al., 2004). Contributions from the regional background and the citywide influence (e.g. high traffic density and industrialization) can lead to widespread chronic exposure, and localized, micro-scale emissions can lead to acute exposure (Turner, 2008). Thus $PM_{2.5}$ measurements, and by association exposure to this hazard, exhibit variability on a range of time scales due to both meteorological variability and variability in emission causing activities (e.g. the hebdomadal (day-of-the-week) cycle). Observation of hebdomadal cycles have been used to quantify the link between anthropogenic emissions and ambient pollution concentrations, primarily in ozone research (Marr and Harley, 2002; Pryor and Steyn, 1995), but variations on hebdomadal time scales have also been observed with PM_{10} (i.e. particles with diameters ≤ 10 μm) (Giri et al., 2006) and black carbon (a constituent of $PM_{2.5}$) (Maciejczyk et al., 2004). For example, decreases in diesel truck emissions of 60–80% during weekends have been linked to decreased elemental carbon concentrations during weekends in California (Harley et al., 2005). Similar weekend traffic reduction was seen in North Carolina, where weekend traffic counts were 18–31% below the 7 day average, and weekend morning rush hour counts were 55–67% below the 7 day average (Perry and Owens, 2001).

Concerns about the degree to which measurements at a small number of fixed locations represent true public exposure (Wilson et al., 2005), combined with recent medical research showing harmful effects of acute exposure to higher concentrations (Brook et al., 2010) and increased understanding of the role of the mobile sector to urban enhancement of PM (and specifically ultrafine particles that may be especially important to health impacts (Fruin et al., 2008)), has led to research on personal exposure of individual

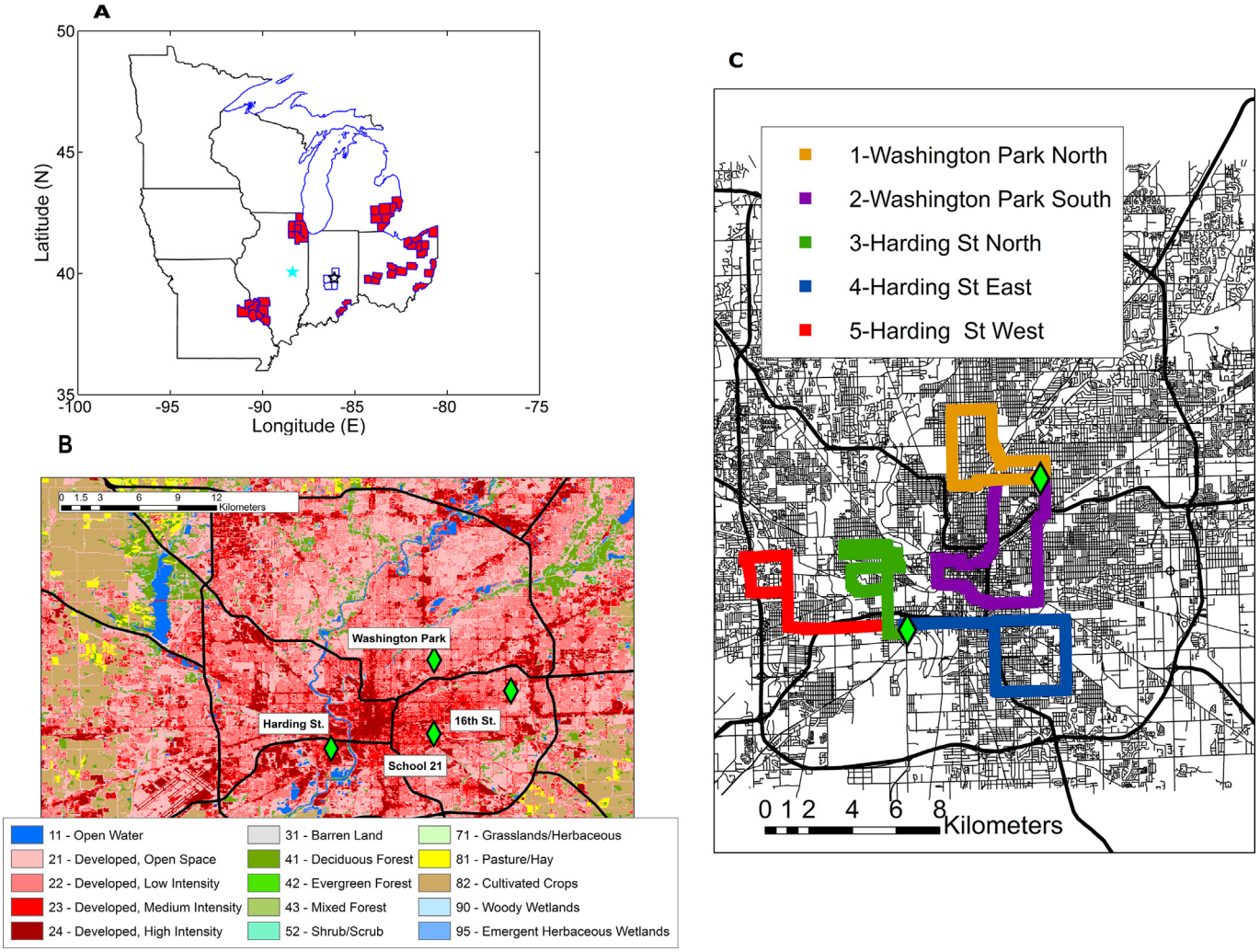

Fig. 1. A) Midwest counties designated as nonattainment for the annual $PM_{2.5}$ NAAQS in 2012 (blue outline) and 2013 (red fill) (data downloaded from http://www.epa.gov/airquality/greenbk/). Also shown is the location of Bondville, IL (cyan star) and Indianapolis, IN (black star). B) Locations of four pDRs in Indianapolis, relative to interstate highways (shown in black). Underlay of land cover demonstrates the variability of level of development in Indianapolis (data downloaded from http://maps.indiana.edu/layerGallery.html?category=LandCover). C) Map of mobile sampling routes starting from the Washington Park and Harding St. sites, where the black lines indicate roadways (data from http://www.census.gov/cgi-bin/geo/shapefiles2013/main). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

subjects or micro-scale emissions on short time scales (Bukowiecki et al., 2002; Gulliver and Briggs, 2004; Kaur et al., 2005).

$PM_{2.5}$ concentrations at any location are an additive function of the regional background (scale; O (>100 km)), local conditions (O (10 km)), and micro-scale emissions (O (<10 km)). An estimate of the regional background $PM_{2.5}$ concentration (and composition) for Indianapolis is derived from the Interagency Monitoring of Protected Visual Environments site located in Bondville, IL (40.052° N, 88.3733° W; Fig. 1a) (Hand et al., 2012). These data indicate similar composition to speciation data from the Indiana Department of Environmental Management (IDEM) site at Washington Park, Indianapolis, IN (one in six day cycle; MET One SASS samplers) from 2005 to 2012 (Fig. 2). At both sites, highest concentrations were observed in summer (July–Sept.), and the lowest concentrations occurred during spring (Apr.–June) and fall (Oct.–Dec.). Sulfate exhibits highest concentrations in summer, and nitrate concentrations are maximized in winter. However, in keeping with previous research indicating urban enhancement of $PM_{2.5}$, on average, $PM_{2.5}$ concentrations in Indianapolis are ~4.5 μg m^{-3} > at Bondville (Fig. 2). The largest difference between $PM_{2.5}$ concentrations in urban Indianapolis and rural Bondville occurs during the summer (5.1 μg m^{-3}) and the smallest difference was observed during winter (3.9 μg m^{-3}), potentially indicating local emission sources make the largest contribution to urban $PM_{2.5}$ during summer.

The aim of this research is to quantify the urban and intra-urban scale $PM_{2.5}$ concentrations and spatiotemporal variability therein using continuous measurements at fixed sites across the city of Indianapolis (Fig. 1b), and the local and micro-scale influences using high temporal resolution mobile sampling (Fig. 1c). The study period presented here (Nov. 2011–Aug. 2013) exhibited daily $PM_{2.5}$ concentrations from the IDEM Federal Reference Method (FRM) measurements that were slightly below the 15 year mean in Indianapolis (Fig. 3; mean $PM_{2.5}$ ~10.9 μg m^{-3} versus 13.8 μg m^{-3}), but the period during which mobile measurements were taken (Aug. 2013) was typical of conditions in Indianapolis (the mean $PM_{2.5}$ = 12.8 μg m^{-3}). Specific research objectives are to:

- Quantify $PM_{2.5}$ variability on diurnal, synoptic, and hebdomadal time scales and determine the dependence on atmospheric processes versus anthropogenic emissions.

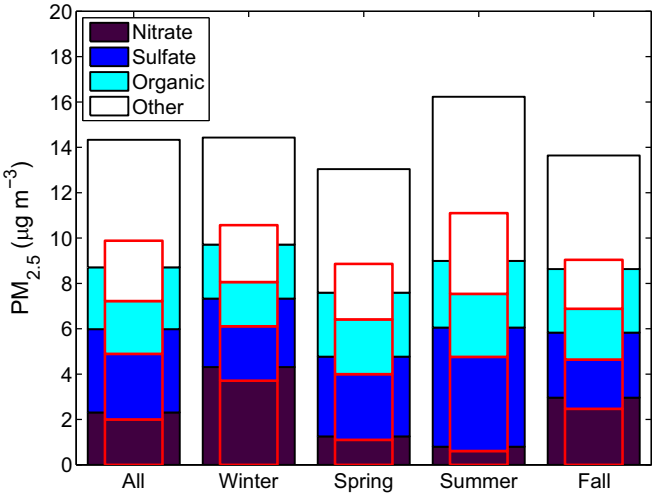

Fig. 2. Mean $PM_{2.5}$ mass speciation at Indianapolis and Bondville (red outline) by season for 2005–2012. See Fig. 1a for the locations of these sites. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

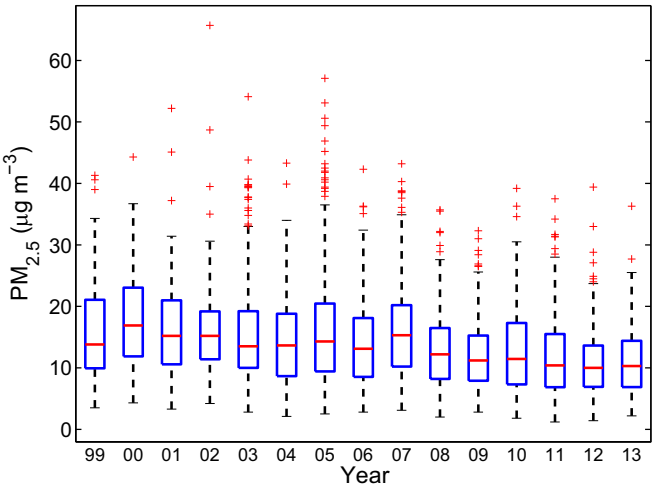

Fig. 3. Annual boxplots of daily mean Federal Reference Method $PM_{2.5}$ concentrations based on measurements at Washington Park, Indianapolis, IN from 7 March 1999–30 Sept. 2013 (data from http://www.epa.gov/airquality/airdata/). Outliers are shown by the red + are lie beyond the 99.3 percentile value. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

- Determine if on short time scales (<1 day), spatial variability is greater than temporal variability in $PM_{2.5}$.
- Quantify spatial variability of $PM_{2.5}$ in proximity to regulatory monitoring sites.
- Quantify spatiotemporal scales of extreme $PM_{2.5}$ concentrations.

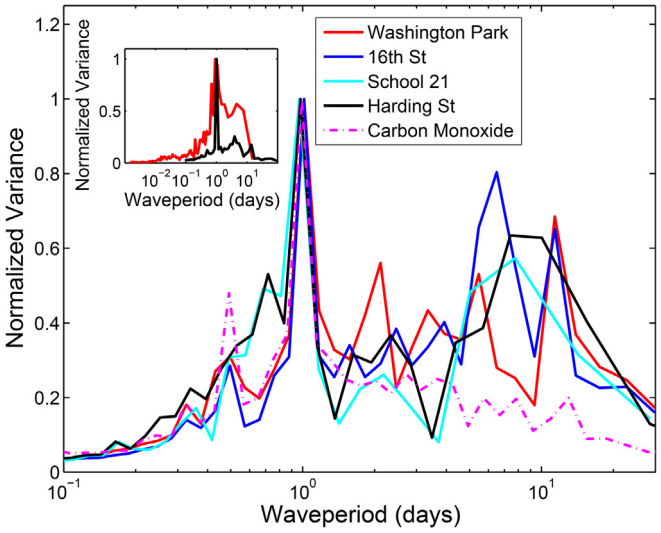

Fig. 4. Normalized variance of hourly averaged $PM_{2.5}$ measurements at Washington Park (11/30/2011–08/25/2013), 16th St. (03/20/2012–08/05/2013), School 21 (03/01/2013–08/05/2013), and Harding St. (03/01/2013–08/24/2013) and carbon monoxide (CO) concentrations at the Washington Park Site. Inset shows 1-min averaged $PM_{2.5}$ measurements (in red) and hourly wind speeds (black) at Washington Park (11/30/2011–08/25/2013). This analysis is based on the fast Fourier transform of the time series wherein missing data periods were filled with the mean concentration from the entire dataset (45%, 35%, 21%, 29% of the time series for Washington Park, 16th St., School 21, and Harding St., respectively) (Wilks, 2006). *Y*-axis indicates the frequency × spectral density at each frequency (i.e. $f \times S(f)$) and is normalized to the maximum value of $f \times S(f)$ at any frequency to facilitate comparison of the frequencies of variability in the CO and $PM_{2.5}$ concentrations. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2. Methods

2.1. Instrumentation and sampling protocols

Ambient $PM_{2.5}$ concentrations were measured using four Thermo Scientific MIE pDR-1500 (hereafter pDR) personal monitors. Although these nephelometric instruments use aerosol optical scattering properties to infer particle concentrations and thus exhibit measurement accuracy which is a function of particle composition, size, and relative humidity (RH), such measurements exhibit relatively high association with particle mass measured using gravimetric filters (Charlson et al., 1969), particularly for $PM_{2.5}$ (Chow, 1995). The pDRs were selected to allow both fixed and mobile measurements. Use of sharp-cut cyclones combined with a flow rate of 1.52 L min^{-1} leads to a 50% aerodynamic cut point of 2.5 μm. For the fixed measurements the pDRs were housed in insulated, temperature controlled enclosures. They were operated without the RH correction (use of the RH correction caused large discrepancies between the units when run in parallel and increased the difference between the pDRs and other $PM_{2.5}$ measurements) and 30-s average measurements were collected.

Prior to deployment the four pDRs were subject to a laboratory inter-comparison. The results indicated relatively good accord. The 30-s $PM_{2.5}$ measurements across a dynamic range of 0–35 μg m^{-3} were used to fit linear regression models which had R^2 values above 0.95 for all instrument combinations, but generally non-zero intercept values of 0.13–0.58 μg m^{-3}. Instrument-to-instrument root mean square errors (RMSE) relative to one of the pDRs across this concentration range were 0.74–1.55 μg m^{-3}. A similar inter-comparison was also conducted at the end of the study deployment. Slight degradation of instrument performance was noted. At the end of the study the R^2 values for 30-s measurements ranged from 0.79 to 0.87, and the RMSE was 1.34–1.89 μg m^{-3}. To provide additional quality control assessment one of the pDRs was run in parallel with gravimetric FRM instruments (operated by IDEM). The daily average $PM_{2.5}$ concentrations ($n = 280$) between the pDR and FRM instruments exhibited an $R^2 = 0.66$, and a RMSE = 5.1 μg m^{-3} (the pDR derived concentration was generally biased high). This analysis thus indicates that the pDRs exhibit relatively high instrument-to-instrument comparability and thus the relative differences in $PM_{2.5}$ concentrations from the units distributed across the city can be seen as robust, but the absolute values must be viewed with caution. Additional testing was conducted to access the degree to which enhanced turbulence and wind speed during mobile sampling influenced the $PM_{2.5}$ concentrations observed using the pDRs. As in prior research (Broich et al., 2012), no systematic difference was observed when the pDRs were operated either by attaching to a person walking or bicycling.

GPS locations during the mobile sampling were measured every 10 s using Garmin GPSMap 78S handheld GPS units (±10 m accuracy).

2.2. Data

2.2.1. Stationary measurements

Stationary measurements were conducted at IDEM monitoring sites (Fig. 1b), to provide a secured location with access to an electrical supply, and integration of data collected by IDEM including carbon monoxide (CO) concentrations (Washington Park and 16th St.) and meteorological conditions (temperature, and wind speed and direction). Continuous 30-sec measurements were conducted at Washington Park (30 November 2011–25 August 2013), 16th St. (20 March 2012–5 August 2013), School 21 (1 March 2013–5 August 2013), and Harding St. (1 March 2013–24 August 2013) at 3 m a.g.l. (~1 m at School 21). These sites are at variable horizontal distances from roadways (Washington Park ~ 30 m; 16th St. ~ 40 m; School 21 ~ 70 m; Harding St. ~ 65 m). $PM_{2.5}$ concentrations did not exhibit consistent variability at frequencies below 1 h (Fig. 4), indicating the sites are not appreciably impacted by highly localized $PM_{2.5}$ emissions, and no important variability is lost by using hourly averaged concentrations in the majority of the following analyses.

2.2.2. Mobile measurements

Mobile sampling with pDRs attached to bicycles was conducted 6–25 August 2013. Based on the U.S. Census Bureau's 2010 census (data from http://www.census.gov/geo/maps-data/data/tiger-data.html), U.S. Geological Survey's 2006 land cover dataset (data from http://maps.indiana.edu/layerGallery.html?category=LandCover), U.S. Environmental Protection Agency's 2008 National Emissions Inventory (data from http://www.epa.gov/ttn/chief/net/2008inventory.html), and the Indiana Department of Transportation's 2011 Annual Average Daily Traffic dataset (data from http://dotmaps.indot.in.gov/apps/trafficcounts/), five routes were designed to transect the urban core, sample population hotspots, and areas where land use is anticipated to contribute to higher $PM_{2.5}$ concentrations. Routes started at a stationary measurement site to allow cross-calibration of the pDR units, and were designed to maximize time in census tracts with population densities >1000 persons km^{-2} with medium to high intensity development, transect the southwest portion of the city where the three largest primary $PM_{2.5}$ point source emissions are located, and cross and/or ride parallel to (<1–2 km from) interstate highways and artillery roads with annual average traffic densities >20,000 vehicles day^{-1} (see Fig. 1c).

pDRs used for the mobile sampling were run in parallel to the unit continuously sampling at the fixed location for ~30 min prior to the transect, for ~30 min between transects (i.e. when the cyclist returned to the start point), and ~30 min after the final transect was completed. During the study, an individual route was sampled multiple times on a given test day. Over the twenty day study period, 3 days had 4 runs, 2 days had 3 runs, 13 days had 2 runs, 1 day had 1 run, and 1 day did not have measurements. All transects were conducted with the first run beginning in the morning hours (median (range) start times of first run and end of last run were 0854 LST (0654–1056 LST) and 1212 LST (1041–1435 LST), respectively). To limit the impact of instrument bias, one instrument was used for all transects, except on days with >2 transects, where another instrument was used for the extra 1–2 runs.

As meteorological conditions were variable over the study period, an adjusted mean concentration was calculated allowing comparison between the transects:

$$\overline{X}^{*} = \overline{X_D}/X_D \times \overline{X} \quad (1)$$

where X_D = daily average $PM_{2.5}$ concentrations and $\overline{X_D}$ = annual average concentration at the stationary site, and $\overline{X}$ = mean of the transect.

To compare how $PM_{2.5}$ concentrations vary on spatial versus temporal scales, the coefficient of variation (COV) was calculated for both the mobile and stationary 30-s $PM_{2.5}$ concentrations:

$$\mathrm{COV} = \sigma/\overline{X} \quad (2)$$

These comparisons were also undertaken excluding transects that had mean $PM_{2.5}$ concentrations < 5 μg m^{-3}, because of the large offset between the pDRs and FRM at low concentrations (for $PM_{2.5}$ < 5 μg m^{-3}: $R^2 < 0.01$ between FRM and pDR; $R^2 = 0.32$, 0.46, and 0.35 between pDRs when run in parallel).

3. Results and discussion

3.1. Stationary measurements

$PM_{2.5}$ concentrations from all four sites exhibit maxima during the early morning hours (~0600 to 0800 LST), and daily minima during the afternoon hours (~1400 to 1600 LST). However, extreme values (defined here as values > 35 μg m^{-3}, ~95th percentile of the hourly average $PM_{2.5}$ concentrations at Washington Park) occurred almost exclusively during the overnight and early morning hours (Fig. 5). This highlights the dual controls of emissions and meteorological conditions, with increased emissions from the mobile sector during morning and evening rush hour, and increased boundary layer heights and enhanced dilution associated with mid-afternoon surface heating. As further evidence of the role of meteorological conditions in determining ambient $PM_{2.5}$ concentrations, hourly observations > 35 μg m^{-3} were associated with lower wind speeds and thus more stagnant conditions (wind speeds were 22–23% lower at all sites when $PM_{2.5}$ > 35 μg m^{-3} than during all sample periods) (Fig. 6a). Wind directions associated with the highest $PM_{2.5}$ concentrations also indicated a higher prevalence of easterly flow at Washington Park (Fig. 6b) and 16th St., and an increased prevalence of southerly flow at Harding St. compared to the average, which may indicate advection from nearby particulate sources (e.g. interstate highways south of Washington Park and east of 16th St., and the largest point source emissions south of Harding St., Fig. 1), although caution should be taken in interpreting these findings as they are possibly manifestations of more stagnant conditions and recirculation in the urban canyons (Vardoulakis et al., 2003).

There is no statistically significant difference in traffic density on urban highways in Indiana on Tuesday through Thursday which is therefore treated as representative of weekday traffic, while traffic volumes were approximately 90% of the 7 day average on Saturdays and 75% of the 7 day average on Sundays (Indiana Department of Transportation Traffic Monitoring Section, 2008). In accord with these data, hebdomadal variability in $PM_{2.5}$ concentrations is also observed in Indianapolis. Upper quantile values are higher during weekdays (Tues–Thurs) compared to weekends (Sat–Sun) at all four sites (Fig. 7). The weekday versus weekend difference for both the 90th and 95th quantile concentrations is statistically significant based on the Wilcoxon rank-pair test (Pryor and Steyn, 1995; Wilks, 2006) ($\alpha = 0.05$) at 16th St. and School 21. These sites are downwind of the major interstate junction in the city's core (Fig. 1b), and thus this finding is consistent with expectations that $PM_{2.5}$ concentrations are generally higher near heavy traffic corridors (Fruin et al., 2008).

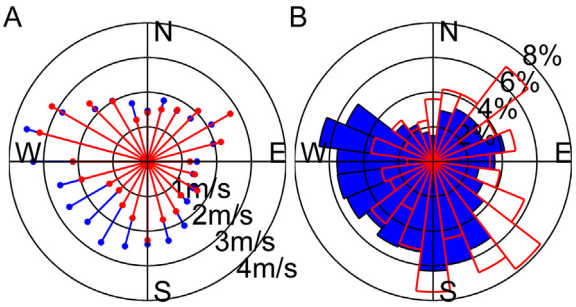

Fig. 6. (A) Mean wind speed by direction and (B) frequency of wind from each direction at Washington Park (11/30/2011–08/25/2013) shown for all measurement periods (blue, $n = 8152$) and during high $PM_{2.5}$ concentrations (>35 μg m^{-3}, shown in red, $n = 334$). (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

Hourly average $PM_{2.5}$ concentrations at the four sites display peaks in variability at multiple frequencies (Fig. 4), including the diurnal time scale (i.e. wave period ~ 10^0 days) that also dominate CO concentrations providing a clear link to primary emissions associated with vehicular traffic (90% of CO emissions in Marion County come from the mobile sector; data from http://www.epa.gov/ttn/chief/net/2011inventory.html (Reynolds et al., 2002)). Data from three of the four sites (Washington Park, School 21, and

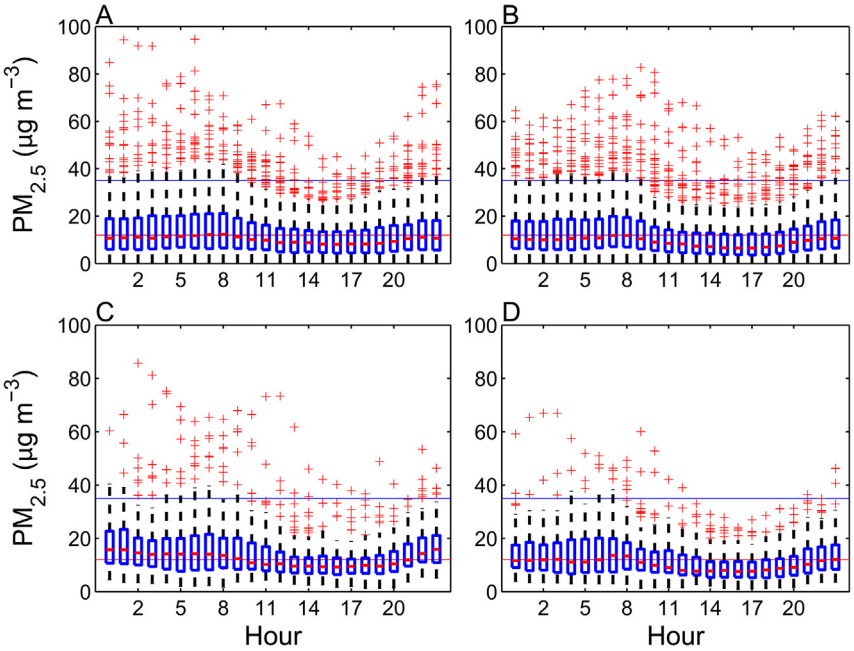

Fig. 5. Boxplots of hourly average $PM_{2.5}$ concentrations by hour of the day (where outliers are approx. > the 99.3 percentile) at A) Washington Park (11/30/2011–08/25/2013), B) 16th St. (03/20/2012–08/05/2013), C) School 21 (03/01/2013–08/05/2013), and D) Harding St. (03/01/2013–08/24/2013). The axis is truncated at 100 μg m^{-3} for legibility (this excludes 2 h at Washington Park and 2 h at School 21). Also shown are the annual (12 μg m^{-3}, red) and daily (35 μg m^{-3}, blue) NAAQS. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

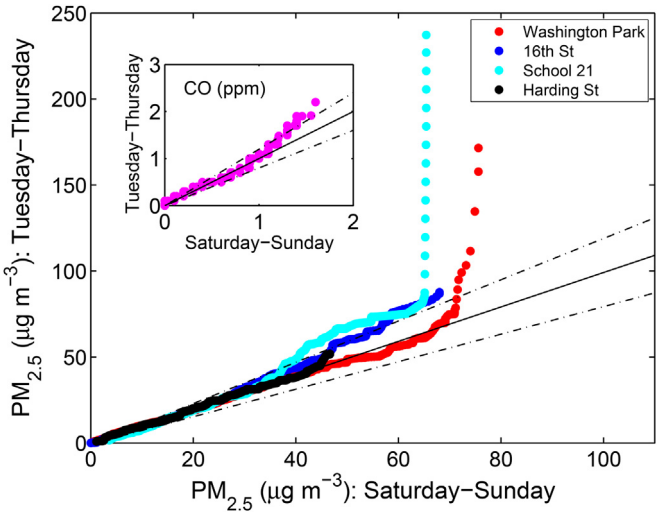

Fig. 7. Empirical Quantile–Quantile plots of $PM_{2.5}$ concentrations and CO (inset) on weekends (Sat–Sun) and weekdays (Tue–Thurs). To aid interpretation also plotted is the 1:1 line (solid), and the dashed lines denote ±20% from the 1:1 (following Pont and Fontan (2001)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Harding St.) also exhibit a secondary maximum at time scales of ~2 days that may be linked to the hebdomadal variability in vehicular traffic. All sites show enhanced variance at time scales of 5–10 days (Fig. 4) which is representative of the time scales of synoptic-scale meteorological phenomena (transitory weather systems) which cause variability in long-range transport, wind speeds (Fig. 4), and the occurrence of precipitation.

Although there are similarities in $PM_{2.5}$ concentrations measured at the four sites (Fig. 5) and the temporal scales of variability (Fig. 4), the Pearson correlation coefficients between hourly average $PM_{2.5}$ concentrations at the sites varied between 0.51 and 0.88. Coefficients of divergence between the sites (CODs, Eq. (3); described in Pinto et al. (2004)) were 0.18–0.42, which is comparable to the least homogeneous metropolitan statistical areas examined in Pinto et al. (2004).

$$\mathrm{COD}_{ab} = \sqrt{1/n \sum_{i=1}^{n} (X_{ai} - X_{bi}/X_{ai} + X_{bi})^2} \quad (3)$$

where n is number of observations, and X_{ai} and X_{bi} are the hourly concentration for hour i at sites a and b, respectively.

$PM_{2.5}$ concentrations above the 90th percentile occurred simultaneously at two sites on 26–89% (mean = 68%) of exceedance hours at an individual site (compared to 13–14% for 1000 randomly generated samples). Thus, while there is coherence in the occurrence of extreme concentrations, on average, 32% of high concentrations measured at one site were not simultaneously measured at other sites across the city. This high degree of spatial variability provided the motivation for the mobile sampling.

3.2. Mobile sampling

A total of 45 individual transects were conducted 6–25 August 2013. The highest mean $PM_{2.5}$ concentrations (Table 1) were observed on the Washington Park South and Harding St. East transects (i.e. transects 2 and 4 in Fig. 1c). The adjusted mean $PM_{2.5}$ concentration (Eq. (1)) on all of the routes that started at Harding St. are higher than the two that originated from Washington Park, while the Harding St. West route (route 5, which was the only route to leave Indianapolis city proper) typically had concentrations 1 μg m^{-3} lower than the other two routes originating at the site (Fig. 1c).

The COV (Eq. (2)) was used to compare spatial versus temporal variability in $PM_{2.5}$ concentrations measured along each transect and in the stationary measurements during the period. On average, the spatial variability is nearly 3 times greater than the temporal variability for all 5 transect routes, with an average mobile COV of 45% compared to 17% for the stationary measurements (Table 1). Even when samples with low mean $PM_{2.5}$ concentrations (<5 μg m^{-3}) are removed, the COV is still 1.78 times higher for the mobile dataset compared to the stationary instruments (COV of 25% and 14%, respectively). Given the importance of mobile emissions to $PM_{2.5}$, at least some of the difference in spatial and temporal variability may be attributable to displacement of the stationary sites from the roadway. Nevertheless, some fraction may also reflect true micro-scale concentration variability (and not only the highly localized impact of vehicles passing the cyclist).

A budget for the contribution of micro-scale emissions was constructed based on the mobile transect sampling as follows. The mean standard deviation (σ) of 30-sec $PM_{2.5}$ concentrations as measured along every transect is 4.2 μg m^{-3}, while the average for individual routes is 2.2–6.0 μg m^{-3} (up to 20.5 μg m^{-3} when individual transect runs are considered). The mean σ of measurements at the fixed sites during the transects is 2.4 μg m^{-3}. Thus assuming the variability at the fixed sampling sites represents advection and the local (i.e. city-wide) addition of $PM_{2.5}$ to the background, we estimate micro-scale emissions along the transects contribute approximately 0–3.6 μg m^{-3} to the $PM_{2.5}$ concentration at the neighborhood scale. These values can be compared to the regional background summertime average daily mean $PM_{2.5}$ of 11.1 μg m^{-3} (Fig. 2).

Data from the mobile transects indicate micro-emissions generate transient periods of extreme concentrations over 200 μg m^{-3} (mean = 4.4 μg m^{-3}, standard deviation = 11.7 μg m^{-3}, Table 1) above local conditions on spatial scales of up to 2 km. To estimate the spatial scales of extremely high $PM_{2.5}$ concentrations, we computed the duration of time with consecutive concentration measurements exceeding one standard deviation above the mean value for the transect (i.e. concentrations > $\overline{X} + 1\sigma$). The results were compared against 'exceedance durations' computed from 300 randomly sampled data sets that had the same mean and standard deviation as each transect to simulate the expected consecutive high concentrations due to stochastic processes. $PM_{2.5}$ concentrations along the transects exhibited coherence above that from stochastic processes for all event durations above 30 s (Fig. 8). Based on an approximate average bicycle speed of 5 m s^{-1} (~11 mi h^{-1}), the mean spatial scale of extreme $PM_{2.5}$ concentrations is 0.5 km, and the range is up to 2 km. Although some 'exceedance durations' lasted >360 s, investigation of those events showed that they are associated with highly non-stationary $PM_{2.5}$ concentrations at the stationary measurement sites (e.g. decrease in $PM_{2.5}$ concentrations associated with enhanced mixing due to rapid boundary layer heating during the transect period).

4. Conclusions

A mixed stationary and mobile sampling design is used to quantify the spatiotemporal variability of $PM_{2.5}$ concentrations in a major urban area, and to undertake diagnostics of possible causes of the variability. High temporal resolution $PM_{2.5}$ measurements at a large metropolitan area in the Midwestern USA indicate variability on both natural (i.e. diurnal and synoptic) and

Table 1
Descriptive statistics of 30-s mean $PM_{2.5}$ concentrations for mobile transect sampling conducted 6–25 August 2013.

Route	Mobile transects								Ratio of mean conc. in mobile transect to stationary measurements	Stationary		
	Mean ($\mu g\ m^{-3}$)	Adjusted mean ($\mu g\ m^{-3}$)	Max ($\mu g\ m^{-3}$)	Min ($\mu g\ m^{-3}$)	σ ($\mu g\ m^{-3}$)	COV	Temp (°C)	Relative humidity (%)		Mean ($\mu g\ m^{-3}$)	σ ($\mu g\ m^{-3}$)	COV ($\mu g\ m^{-3}$)
1	14.13	11.74	67.44	3.82	2.16	0.18	26.63	46.84	2.62	7.97	0.81	0.15
2	17.31	12.20	300.72	2.26	5.75	0.29	28.38	46.55	0.90	20.42	4.08	0.13
3	15.87	13.74	30.94	0.46	2.37	0.25	28.14	41.81	1.20	11.14	1.40	0.13
4	17.21	13.99	80.31	−0.36	4.83	0.60	27.36	40.43	1.25	14.22	3.23	0.23
5	15.21	12.55	111.05	−1.30	6.03	0.91	28.14	41.81	1.42	14.00	2.45	0.21

Stationary statistics represent the $PM_{2.5}$ concentrations measured at the starting site (fixed measurement location) during the time period of each transect. Routes are shown in Fig. 1c.

anthropogenic (i.e. diurnal and hebdomadal) cycles. This indicates a link between $PM_{2.5}$ concentrations and meteorological conditions with the greatest variability occurring on diurnal time scales and higher concentrations being associated with stagnant conditions. Higher extreme concentrations occur on weekdays and similarities in the frequencies at which $PM_{2.5}$ and CO exhibit maximum variability indicate a link to mobile sector emissions. This finding is consistent with prior research which has indicated the mobile sector as a key determinant of urban enhancement of $PM_{2.5}$, and indicates that implementation of state-wide vehicle emissions testing (such as has been implemented in many California counties; Maga and Hass (1960)) might be a plausible step in reducing the health burden of $PM_{2.5}$ on urban residents.

Mobile measurements taken by deploying the pDRs on bicycles indicate that spatial variability is as much as 2–3 times greater than temporal variability on sub-daily time scales. Local differences in $PM_{2.5}$ concentrations along the transects (which are <25 km in length) are of comparable magnitude to the observed difference between stationary measurements in the urban environment relative to a regionally representative background station. This finding emphasizes a key challenge in relating city-wide total mortality to a representative $PM_{2.5}$ concentration in epidemiological studies (Pinto et al., 2004).

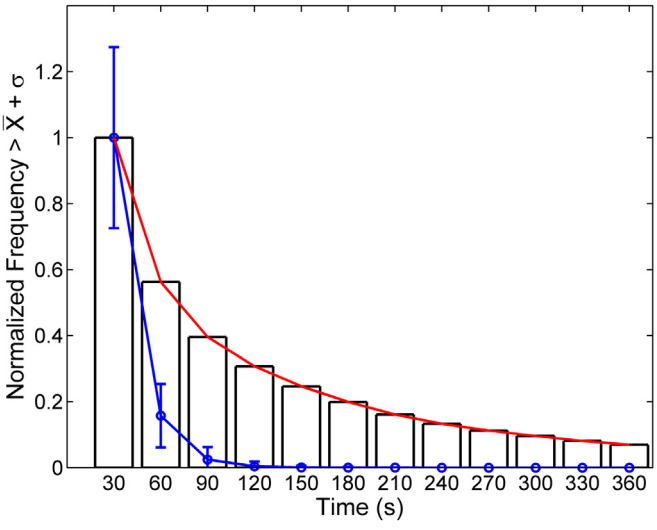

Fig. 8. Histogram of the exceedance duration (i.e. the duration of time in seconds, where $PM_{2.5}$ concentrations exceeded the transect mean plus one standard deviation), normalized by the number of excursions in the sample (red line and bars). Also shown in the blue line is results from 300 randomly generated samples per transect (representing exceedances from purely stochastic processes). The vertical bars denote ±one standard deviation. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

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