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Passive sampling to capture spatial variability in $PM_{10-2.5}$

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Abstract

This work applied inexpensive passive sampling to measure airborne coarse particles with aerodynamic diameters between 2.5 and 10 μm ($PM_{10-2.5}$) over three 7-day periods at 33 sites in a medium-sized Midwest City (Iowa City, IA). The number of sites and their locations were selected using an optimal sampling design that captured 95% of the total variance in $PM_{10-2.5}$ as measured with real-time sampling equipment on a mobile sampling platform. Weekly averages of $PM_{10-2.5}$ were 15.9 $\mu g\,m^{-3}$ (coefficient of variation between sites, CV = 23%), 17.9 $\mu g\,m^{-3}$ (CV = 24%), and 6.1 $\mu g\,m^{-3}$ (CV = 30%). ANOVA showed that these means were statistically different ($p<0.001$), and that the spatial variability plus random error accounted for 29% of the total variability. The maximum coefficient of divergence between sites—a relative measure of uniformity—ranged from 0.21 to 0.36. These values indicate that $PM_{10-2.5}$ was heterogeneous even on the fine spatial resolution studied in this work (average distance between sites was 4.4 km). The spatial patterns of $PM_{10-2.5}$ measured with the passive samplers closely matched with those of mobile mapping and corresponded with known coarse particle sources in the area. This work demonstrates that passive sampling coupled with effective sampling design may enhance our ability to assess exposure to $PM_{10-2.5}$ at a local scale. Compared to exposure estimates made with data from centrally located, filter-based samplers, these highly spatially resolved estimates should reduce exposure misclassification errors in epidemiological studies.

Keywords: Exposure assessment; Coarse particles; Spatial variability; $PM_{10-2.5}$; Particulate matter

1. Introduction

Airborne particulate matter has been associated with adverse cardiopulmonary health effects (Krewski and Rainham, 2007). The type and severity of such health effects depend on the size, concentration, and composition of particles that a person inhales (Hinds, 1999). Consequently, the US EPA regulates particulate matter by particle size as $PM_{2.5}$ defined as the mass concentration of "fine" particles that are <2.5 μm in aerodynamic diameter, and PM_{10} defined as the mass concentration of particles with an aerodynamic diameter <10 μm (USEPA, 2004). In 2006, the US EPA proposed to replace the PM_{10} standard with $PM_{10-2.5}$ or "coarse" particles, defined as the difference between PM_{10} and $PM_{2.5}$ (USEPA, 2006b). This change would not only have eliminated the duplication in regulation of particles <2.5 μm in both PM_{10} and $PM_{2.5}$ but also have been preventative against adverse health effects that may only be associated

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with exposure to ambient coarse particles. The US EPA ultimately maintained the PM_{10} standard, citing the lack of conclusive evidence regarding the association of adverse health effects with exposure to $PM_{10-2.5}$ (USEPA, 2006a).

The epidemiological evidence that links adverse health effects to coarse particle exposure is substantially weaker than that for fine particles (Brunekreef and Forsberg, 2005), although this fact may stem from limitations of exposure assessment rather than a true lack of effect (Wilson et al., 2005). Most epidemiological studies rely on data from compliance networks composed of sparsely distributed filter-based samplers (USEPA, 2004). Such estimates may be adequate when particulate matter is fairly homogeneous, such as accumulation mode aerosol (0.1–1 μm) that usually dominates $PM_{2.5}$ and may contribute substantially to PM_{10} in urban settings (Wilson et al., 2005). $PM_{10-2.5}$, however, may vary substantially across time and space because the largest particles that compose this size fraction settle by gravity rapidly as they move away from a source (Hinds, 1999). Thus, reliance on central monitoring as a proxy for personal exposure to $PM_{10-2.5}$ may result in considerable exposure misclassification as compared to that for $PM_{2.5}$ (Brunekreef and Forsberg, 2005; Monn, 2001; Wilson et al., 2005). Exposure misclassification may alter the significance, direction, and magnitude of associations found between particle exposure and adverse health outcomes (Ito et al., 2004).

Substantial intra-city variations have been observed in the sources, size, and composition of airborne particles (Henderson et al., 2007; Jerrett et al., 2007; Ross et al., 2006). In a meta study by Wilson et al. (2005), PM was found to be heterogeneous in: 100% ($n = 4$) of studies for $PM_{10-2.5}$, 60% ($n = 20$) for PM_{10}, and 43% ($n = 21$) for $PM_{2.5}$. The US EPA made similar observations from analysis of data from compliance networks in 17 US cities (USEPA, 2004). However, these analyses provide little insight into the spatial variability in $PM_{10-2.5}$ at a local scale ($\leqslant$10 km) because the estimates of each city are based only on several samplers that are spread over large distances (10–100 km). Knowledge of the variability at a local scale, however, is critically important to avoid exposure misclassification, and the expense of purchasing and operating a sufficient number of filter-based samplers to capture this variability is often impractical.

Thus, the overall goal of this work was to use novel sampling methods to derive estimates of the spatial variability in $PM_{10-2.5}$ on a local scale. Passive samplers were deployed at 33 sites across a medium-sized Midwest City. The number of sites and their locations were selected to capture 95% of the total variance in $PM_{10-2.5}$ as measured with real-time samplers mounted on a mobile platform. The results of this work are likely to help us understand the magnitude of exposure misclassification when data from central monitoring sites are applied to estimate exposure to $PM_{10-2.5}$. In addition, this study demonstrates the effectiveness of passive sampling coupled with effective sampling design through mobile sampling as a potential solution to improve exposure estimates for coarse particles at a local scale.

2. Methods and materials

2.1. Preliminary mapping and site selection

The number of $PM_{10-2.5}$ monitoring sites and their locations were selected with a novel procedure to optimize spatial sampling for exposure assessment. First, a preliminary estimate of the variability in $PM_{10-2.5}$ was obtained through real-time mapping. $PM_{10-2.5}$ was measured with data from an optical particle counter (OPC; PDM-1108, Grimm, Ainring, Germany) that was loaded into the rear section of a minivan. Each sample day all equipment was turned on and allowed to warm up for a minimum of 15 min prior to sampling. The minivan was then driven to the first site, the engine was turned off, the rear of the van was opened, and any exhaust or dust resuspended from driving was allowed to dissipate for 2 min. Particle concentrations were measured with the OPC for 6 min, averaged, and logged to a data file onto a laptop computer. The minivan was then driven to the next site and the process was repeated. Over a 4-day period in the summer of 2006, 171 measurements of $PM_{10-2.5}$ were made at 67 locations across Iowa City and its surroundings. These data were used to generate a surface of $PM_{10-2.5}$ (Fig. 1) using Kriging method described by Cressie (1990). From basic statistics, the number of sample sites, n, needed to capture 95% of the observed variability was calculated as

$$n = \left(\frac{1.96s}{U}\right)^2, \quad (1)$$

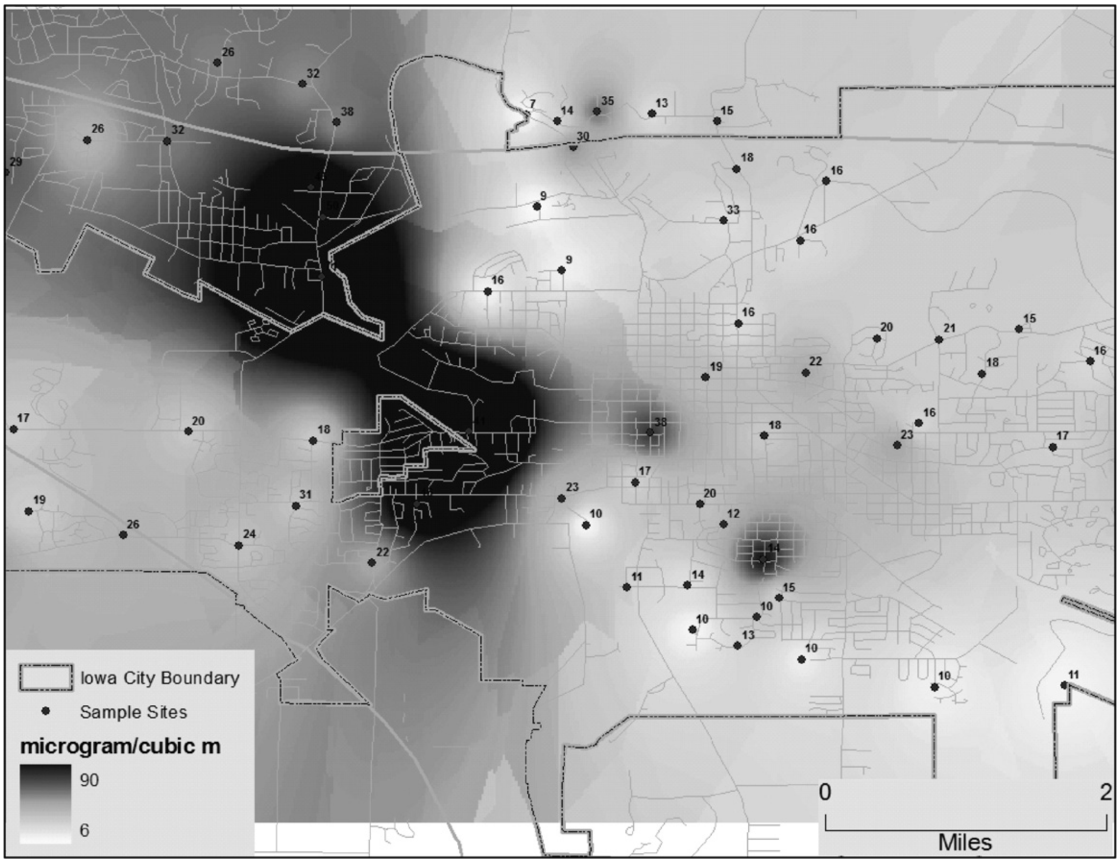

Fig. 1. Distribution mean $PM_{10-2.5}$ from mobile sampling.

where s is the sample standard deviation and U is the acceptable uncertainty (5% of the range of $PM_{10-2.5}$ observed in mobile sampling). Using the results from mobile sampling $s \sim 9.32\,\mu g\,m^{-3}$ and $U \sim 3.18\,\mu g\,m^{-3}$, n was estimated as 33 sites.

The designated sample area was a 112.5 km^2 (12.25 km × 9.2 km) centered on the city. The extent of the area was −91.462764° to −91.609674° longitude and 41.617258° to 41.699758° latitude (Fig. 1). A grid of 56 rows and 99 columns was overlaid onto the $PM_{10-2.5}$ surface, which resulted in a total of 5544 cells (N). A novel procedure was employed to choose 33 sites from the 5544 candidates by maximizing variance and minimizing spatial autocorrelation in $PM_{10-2.5}$ as

$$\max(Z) = \sum_{i=1}^{n} \sum_{j, j \neq i, d_{ij} \geqslant h}^{n-i} (z_i - z_j)^2 d_{ij}, \tag{2}$$

where z_i and z_j represent $PM_{10-2.5}$ measured at ith sample site and its jth neighbor, respectively, d_{ij} is the distance between ith sample site and its jth neighbor, and h is the range to avoid spatial autocorrelation. The range is the distance beyond which spatial autocorrelation becomes insignificant and was equal to 0.015° computed with the aid of empirical semivariogram of $PM_{10-2.5}$ data from mobile sampling. The adequacy of the optimal sample design was determined using an empirical semivariogram of the data collected at the 33 sample sites.

Fig. 2 depicts the 33 site locations that were selected on a street map of the city. Interstate 80 and The Old Capitol Building (identifying downtown Iowa City) are highlighted to provide a frame of reference on the map, and potential sources of $PM_{10-2.5}$ are indicated with a circle. Four sources (two large rock quarries, a major construction site, and an industrial area) in the northwest quadrant were technically outside of the city limits but were thought to potentially affect air quality. The other three sources identified were a large construction

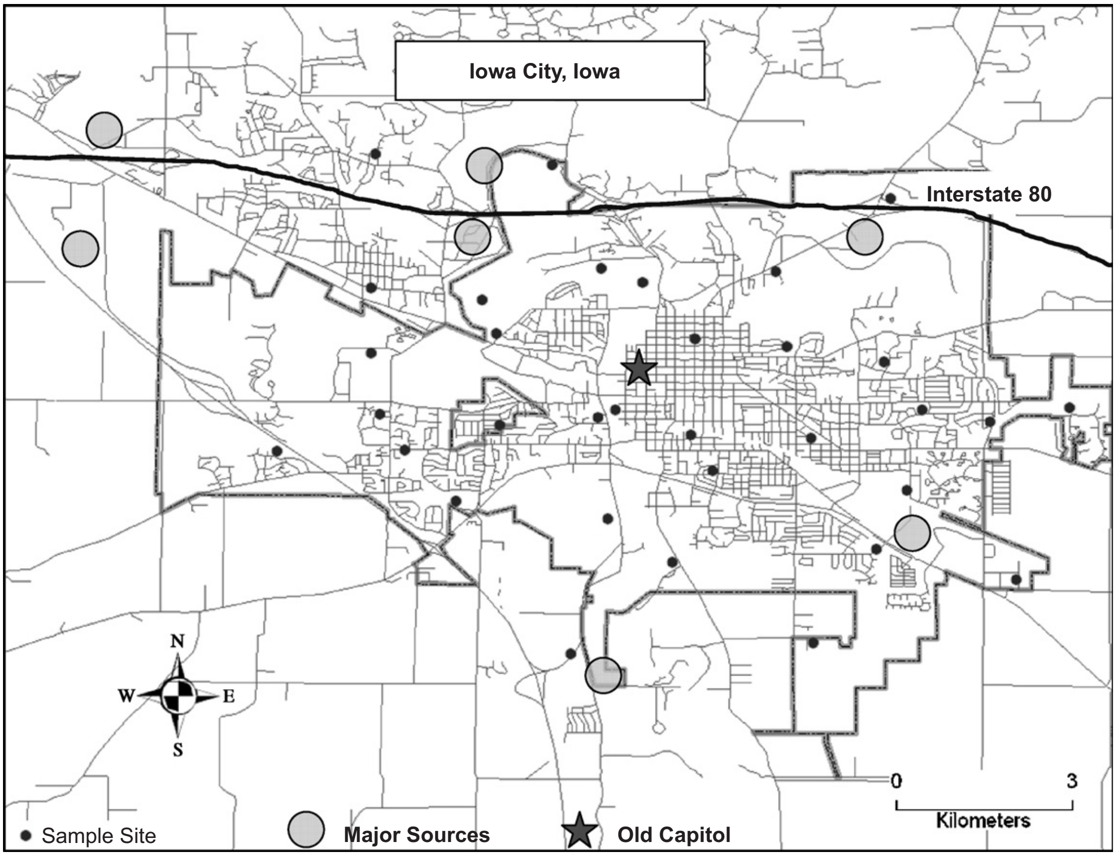

Fig. 2. Street map of Iowa City with sample sites. Major sources include quarries, industrial areas, and major construction sites operating during the sample period.

site (northeast quadrant), an industrial area with moderate road construction also occurring in the area (extreme southeast), and an area with several factories, a small quarry, and road construction activities in close proximity of each other (south central).

2.2. Passive sampling

Wagner and Leith (2001a, b) introduced a passive sampler to measure particles indoors. In this sampler, particles deposit by diffusion, gravity settling, and turbulent inertial forces onto a collection substrate. They developed a deposition velocity model to translate particle surface loading on the substrate, determined through microscopy, to airborne mass concentration. In previous work, we developed a shelter to protect this passive sampler from the effects of wind and precipitation for ambient deployment (Ott, 2007 and Ott and Peters, submitted for publication).

Passive samplers installed in shelters were deployed at each site for three 7-day periods during consecutive weeks in December 2006. The shelter was placed on top of its own support post at 1.8 m above the ground. Samplers were placed away from the immediate vicinity (>30 m) from obvious particle sources such as a busy road because measurements were intended to represent exposures over the broad area in which they were placed.

All samplers were delivered to the site and started on the first day of the sample period. An identical schedule was followed each week so that samplers started and stopped at nearly the same time of the day for a given site. The delivery and change-out of samplers took about 6 h for all the 33 sites. Although there were slight differences in start and stop times among the samplers, the difference was a maximum of 6 h or <3.6% of the 7-day sample period. At two sites, SW12 and NE33, meteorological equipment (Weather Transmitter WXT510, Vaisala, Helsinki, Finland or Oregon Scientific

WMR968, Portland, OR) was used to record temperature, relative humidity, rainfall, and wind speed within 2 m of and at the same height as the passive samplers. Three blank passive samplers were transported and analyzed for each period.

2.3. Passive sampler analysis

Passive samplers were analyzed as discussed in detail by Ott (2007) and Ott and Peters (submitted for publication). Digital images of the particles on the sampler's glass media were captured at 100 × magnification (10 × objective lens) with a light microscope (Leica DMLSP, Leica Microsystems, Wetzlar, Germany) equipped with a digital camera (Leica DFC 280, Leica Microsystems). Particles were counted and sized automatically with ImageJ (NIH, Bethesda, MD). This imaging process was adjusted so that the size of 5-μm borosilicate glass microspheres (Catalog # 9005, Duke Scientific, Fremont, CA) indicated by ImageJ was within 5% of their NIST-certified diameter.

Particle mass was computed by assuming that aerodynamic diameter was equal to the projected area diameter from microscopy, d_{pa}. The mass of a single particle, m_i, was then calculated using

$$m_i = \left(\frac{\pi}{6}\right)\rho_p\left(\frac{d_{pa}}{S_V}\right)^3, \tag{3}$$

where ρ_p is the density of the particle and S_V is the volumetric shape factor. Particle density was assumed to be 2.0 g cm^{-3} following Wagner and Macher (2003). S_V was estimated from particle circularity, C_p, output by ImageJ as: $S_V = 1/C_p$.

The contribution of a single particle to airborne mass concentration, C_i, was calculated as

$$C_i = \frac{F_i}{V_{dep,i}} = \left(\frac{m_i}{A_T t}\right)\frac{1}{V_{dep,i}}, \tag{4}$$

where F is the mass flux of the particle to the deposition surface, A_T is the total area of the sample that was imaged (A_T = number of images times the area of one image), and t is the sample time. $V_{dep,i}$ is the deposition velocity of the particle adjusted by an empirically derived factor accounting for the behavior of the sampler as described by Wagner and Leith (2001a).

$PM_{10-2.5}$ was then calculated as

$$PM_{10-2.5} = \sum_{i=1}^{n} C_i E \quad (\text{if } d_{pa,i} > 2.5\,\mu\text{m}), \tag{5}$$

where E is the PM_{10} curve calculated as (Hinds, 1999)

$$E_i = 0.9585 - 0.00408 d_{pa}^2. \tag{6}$$

A $PM_{2.5}$ curve was not included in the calculation because particles $<2.5\,\mu$m could not be sized accurately. Airborne mass concentration from blank passive samplers was subtracted from the corresponding samples. This subtraction accounted for 6.8% of the total mass on average.

2.4. Data analysis

Pearson correlation coefficient, r, and coefficient of divergence (COD) were calculated for the relationship between $PM_{10-2.5}$ measured with the passive sampler at each site and all other sites (528 comparisons for each site, Excel, Microsoft Corp., Redmond, WA). COD was calculated following Wongphatarakul et al. (1998):

$$COD_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}\left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}}\right)^2}, \tag{7}$$

where x_{ij} and x_{jk} represent the 7-day $PM_{10-2.5}$ for week i at sampling site j, k is the number of sites, and p is the number of observations. We present the minimum r-value and the maximum COD value for each site because they reflect the level of heterogeneity in $PM_{10-2.5}$.

ANOVA was performed to test the hypothesis that the mean of the three weekly averages were equal given the variation within each week. It also allowed a comparison of the within-site to between-site variability. Within-week variability is related to spatial variability among sites, while between-week variability indicated the temporal or weekly variation.

3. Results

Table 1 summarizes $PM_{10-2.5}$ measured with passive samplers. Mean $PM_{10-2.5}$ was similar during Week 1 (15.9 μg m^{-3}) and Week 2 (17.9 μg m^{-3}) when rainfall was minimal (<0.01 cm day^{-1}). However, it was substantially lower during Week 3 (6.1 μg m^{-3}) when rain occurred (0.65 cm day^{-1}). ANOVA confirmed that the three weeks had different mean values ($p<0.0001$). The between-site variation plus random error accounted for 29% of the total variability. The coefficient of variation (CV) of $PM_{10-2.5}$ measured with the 33 passive samplers was similar for the 3 weeks and ranged from 23.1% to 29.2%. The grand mean of $PM_{10-2.5}$ for the 3 weeks was $13.2\,\mu g\,m^{-3}$ with a CV of 46.4%. All samples exceeded the limit of detection of the passive sampler for a 7-day period of $1.7\,\mu g\,m^{-3}$ (Ott, 2007 and Ott et al., submitted for publication). One sample out of 99 was not analyzed because of malfunction (Week 2, sample site SE3) and therefore was excluded from the analysis.

Table 1
Summary of passive sampling

	Week 1	Week 2	Week 3	Overall
Mean $PM_{10-2.5}$ ($\mu g\,m^{-3}$)	15.9	17.9	6.1	13.2
CV (%)	23.1	23.3	29.2	19.8
Minimum $PM_{10-2.5}$ ($\mu g\,m^{-3}$)	9.7	11.5	3.3	9.3
Maximum $PM_{10-2.5}$ ($\mu g\,m^{-3}$)	26.1	31.6	11.1	20.1
N	33	32	33	98
Rain (cm day^{-1})	0.01	0.00	0.65	–
Temperature (°C)	−1.1	1.7	0.8	–
Relative humidity (%)	71.5	70.1	84.5	–
Wind speed (km h^{-1})	9.3	6.0	5.9	–

Table 2 provides mean $PM_{10-2.5}$, CV, locating coordinates, minimum r, and maximum COD by site. A CV near zero, a minimum r near one, and a maximum COD near zero would indicate homogeneity in $PM_{10-2.5}$, while greater heterogeneity in $PM_{10-2.5}$ is indicated as CV value becomes larger, minimum r-value approaches zero, and maximum COD approach one. Wilson et al. (2005) proposed a criterion of CV > 20% to indicate heterogeneity. The US EPA proposed a criterion of COD > 0.2 to indicate heterogeneity and a COD < 0.1 to indicate

Table 2
Sites with coordinates and summary results[a]

No.	Site	Latitude	Longitude	Mean $PM_{10-2.5}$	CV (%)	r (minimum)	COD (maximum)
1	SE11	41.653777	−91.481544	14.3	41.3	0.73	0.23
2	SE10	41.655925	−91.469330	13.1	33.7	0.81	0.24
3	NE34	41.655466	−91.491881	12.0	41.3	0.60	0.24
4	NE33	41.662923	−91.497831	13.1	55.8	0.76	0.23
5	SE6	41.651208	−91.508938	12.3	46.3	0.80	0.23
6	SE7	41.643171	−91.494186	15.1	36.5	**0.37 L**	0.30
7	SE9	41.629435	−91.477525	14.2	47.5	0.65	**0.21 L**
8	SE8	41.634172	−91.498967	12.1	48.1	0.77	0.24
9	SE4	41.619745	−91.508629	**9.3 L**	47.7	0.61	**0.36 H**
10	SE3	41.632144	−91.530395	13.3	**23.4 L**	0.65	0.31
11	SE5	41.646150	−91.524167	13.9	46.5	0.62	0.22
12	SE2	41.651665	−91.527443	12.6	49.8	**0.83 H**	0.23
13	SE1	41.655520	−91.538995	20.1	50.6	0.82	**0.36 H**
14	NE29	41.666521	−91.526861	9.5	32.0	0.56	0.34
15	NE32	41.665234	−91.512767	10.4	52.3	0.51	0.33
16	NE30	41.676824	−91.514357	10.8	49.0	0.73	0.30
17	NE31	41.688107	−91.496845	11.3	49.4	0.61	0.28
18	NE28	41.693307	−91.548764	17.1	62.1	0.55	0.30
19	NE27	41.672511	−91.559518	15.5	49.2	0.81	0.25
20	NW26	41.677412	−91.541274	11.9	55.0	0.66	0.27
21	NW25	41.675144	−91.534768	11.6	52.5	0.82	0.27
22	NW24	41.667323	−91.557255	15.0	51.0	0.82	0.23
23	SW13	41.654403	−91.541774	12.5	49.8	0.80	0.23
24	SW12	41.638764	−91.540162	12.7	48.8	0.77	0.22
25	SW14	41.617971	−91.545966	12.8	50.6	0.60	0.22
26	SW15	41.641471	−91.563576	12.7	48.7	0.81	0.22
27	SW19	41.653238	−91.556791	**20.1 H**	57.6	0.54	0.35
28	SW17	41.649371	−91.571412	11.1	60.8	0.80	0.33
29	SW16	41.649161	−91.591072	11.3	57.8	**0.37 L**	0.32
30	SW18	41.654921	−91.575260	17.7	44.5	0.68	0.32
31	NW20	41.664248	−91.576580	10.9	52.5	0.82	0.30
32	NW22	41.674391	−91.576580	15.6	56.9	0.49	0.24
33	NW23	41.694917	−91.575844	11.9	**64.7 H**	0.73	0.32

[a]Low (L) and high (H) values are shown in bold.

homogeneity of concentration (USEPA, 2004). In the present study, CV values ranged from 23.4% to 64.7%, minimum r-values ranged from 0.37 to 0.83, maximum COD values ranged from 0.21 to 0.36.

Fig. 3A displays the individual weekly $PM_{10–2.5}$ by site, and Fig. 3B shows the mean $PM_{10–2.5}$ for the three 7-day period by site. The x-axis of these plots (the "No." column in Table 3) is the order that samplers were placed at the sites in a given day. $PM_{10–2.5}$ was substantially lower over all sites during Week 3 than Week 1 or Week 2. However, those sites with the lowest or greatest $PM_{10–2.5}$ values were generally consistent between weeks. For example, $PM_{10–2.5}$ observed at Site 27 (SW19) was greater than that observed at most other sites, and $PM_{10–2.5}$ observed at Site 28 (SW17) was generally lower than that observed at other sites.

Fig. 4 is a semivariogram that shows average semivariance on the y-axis against the distance between sample sites along the x-axis. A gradual increase in average semivariance with distance would have indicated the presence of spatial autocorrelation. The lack of relationship between distance and average semivariance indicated that the data were independent and that spatial autocorrelation was avoided.

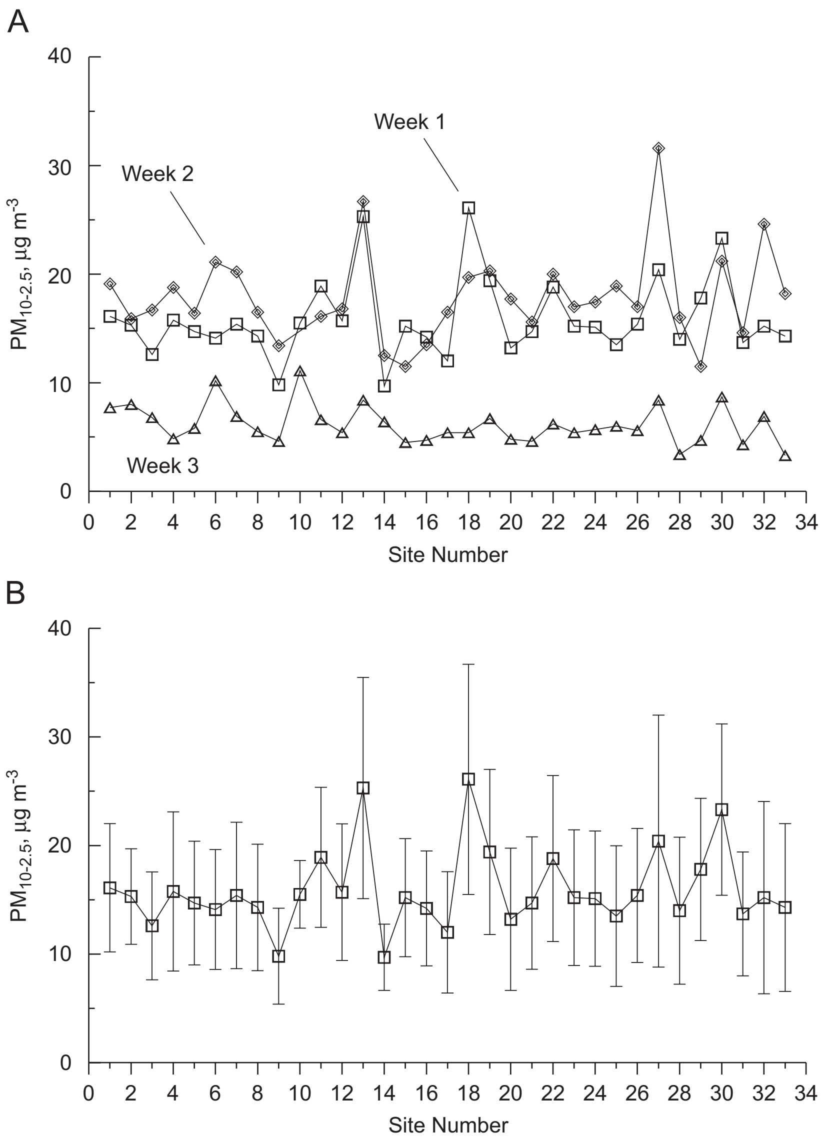

Fig. 3. $PM_{10–2.5}$ by site. Graph (A) is the individual results for each week. Graph (B) is the average of the three runs.

Table 3
Summary of EPA AIRS $PM_{10-2.5}$ intra-urban variability from 2004 Particulate Matter Air Quality Criteria Document

	City	No. of sites	R (minimum–maximum)	COD (minimum–maximum)
1	Columbia	2	0.70	0.37
2	Tampa	2	0.81	0.17
3	Cleveland	6	0.22–0.74	0.18–0.62
4	Steubenville[a]	4	0.48–0.69	0.77
5	Detroit	3	0.39–0.58	0.54–0.79
6	Milwaukee	2	0.65	0.53
7	Chicago[a]	3	0.53–0.82	0.40
8	Gary[a]	3	0.60–0.79	0.83
9	Louisville	2	0.65	0.48
10	St. Louis	3	0.70–0.82	0.76–0.91
11	Baton Rouge	2	0.4	0.43
12	Dallas	4	0.60–0.79	0.17–0.32
13	Salt Lake City[a]	3	0.70–0.74	0.47
14	Portland[a]	2	0.69	–
15	LA	4	0.54–0.82	0.18–0.27
16	Riverside	4	0.32–0.80	0.28–0.39
17	San Diego	4	0.01–0.70	0.25–0.45

This analysis was based on 1 year of 24-h $PM_{10-2.5}$ measured with federal equivalent method samplers. The distance between sites generally ranged from 10 to 40 km.
[a]Not all pairs had a COD reported.

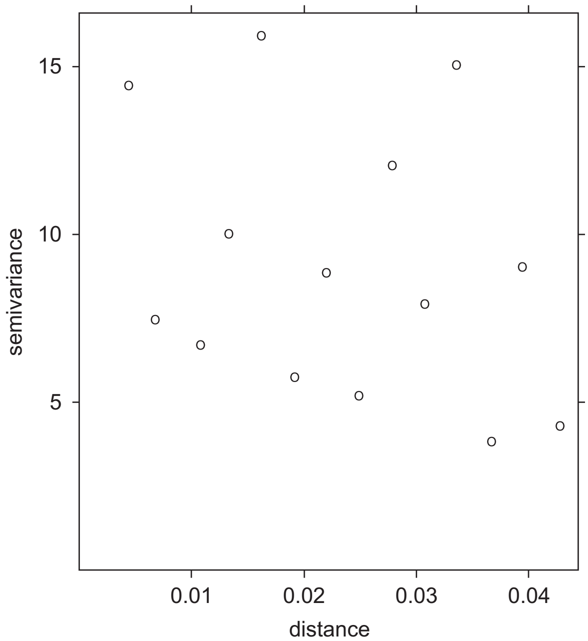

Fig. 4. Semivariogram of mean $PM_{10-2.5}$ difference versus distance between sites.

Fig. 5 shows mean $PM_{10-2.5}$ overlaid on a street map of the city. The $PM_{10-2.5}$ surface was calculated by the inverse distance weighting method, exponent = 1.25 (Shepard, 1968). The greatest values of $PM_{10-2.5}$ were downwind (the prevailing wind direction is from the northwest) of the cluster of four coarse particle sources in the northwest quadrant. The high and low values of $PM_{10-2.5}$ near each other provide further evidence of a lack of spatial autocorrelation. The distribution of $PM_{10-2.5}$ is similar to that obtained through mobile sampling (Fig. 1).

4. Discussion

This work demonstrates the effectiveness of passive sampling coupled with an optimal sampling design to assess the spatial variability in $PM_{10-2.5}$ at a local scale. Passive samplers were deployed at 33 sites across a medium-sized Midwest City. The number of sites and their locations were selected to capture 95% of the total variance in $PM_{10-2.5}$ as measured with real-time sampling equipment on a mobile sampling platform.

Analyses of data from passive samplers suggest that $PM_{10-2.5}$ was heterogeneously dispersed throughout the study area. The weekly CV ($23\% \leqslant CV \leqslant 29\%$) was greater than the criterion of 20% to indicate heterogeneity. The value of maximum COD ranged from 0.21 to 0.36, greater than the criterion suggested by the US EPA to indicate heterogeneity ($COD > 0.2$).

The values of maximum COD observed in this work, however, were lower than those from

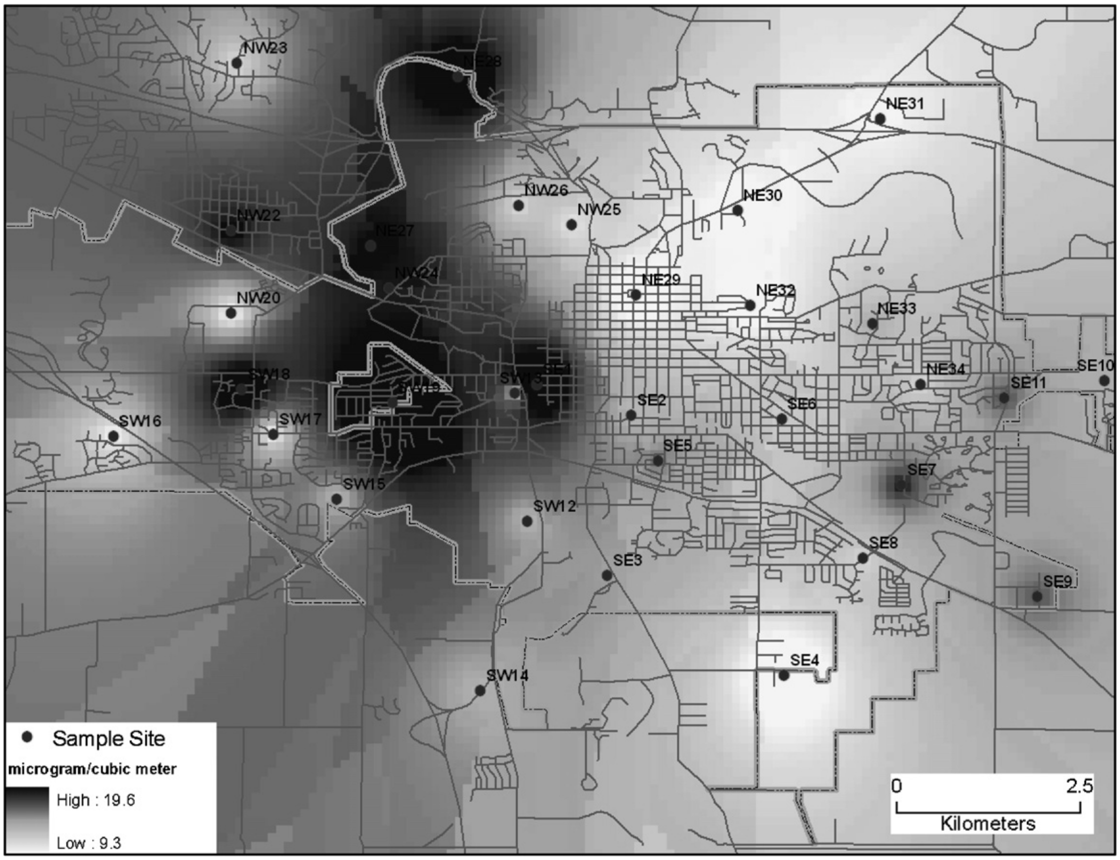

Fig. 5. Distribution of mean $PM_{10-2.5}$ in Iowa City.

compliance monitoring networks (Table 3, USEPA, 2004): values of maximum COD ranged from 0.17 to 0.91 in the 17 cities where $PM_{10-2.5}$ was measured at multiple sites. Lower values of COD would be expected when sample sites are closer (average distance between sites was 4.4 km in this study and 10–40 km in compliance networks) and when the averaging time is longer (averaging time was 7 days in this study and 24 h in compliance data). They would also be expected when the number of samples is lower (only three samples were included in the present study but a year of data was included in the analysis of compliance data). These results point to an inherent limitation in comparing COD values when distance between sites, averaging time, and sample number are not standardized.

The general trend of coarse particles within the study area was consistent across seasons. Although the sampling seasons and durations of passive and mobile samplings were different, the general spatial distribution of coarse particles observed with passive sampling (conducted during 3 weeks in December, Fig. 5) agreed well with that observed with mobile sampling (conducted during 4 days in August, Fig. 1). The greatest values of $PM_{10-2.5}$ were downwind (the prevailing wind direction is from the northwest) of the cluster of four coarse particle sources in the northwest quadrant (Fig. 2). Of these four potential sources (two large rock quarries, a major construction site, and an industrial area), the two large rock quarries may be expected to be the most consistent source of $PM_{10-2.5}$ in the area across seasons. Further sampling with real-time instruments coupled with meteorological equipment will be needed to confirm this observation.

Other areas of elevated $PM_{10-2.5}$ were limited to a single site that was inconsistent spatially between the mobile and passive samplings and was not associated with potential coarse particle sources in the area. These areas may be impacted by local construction that differed across seasons. Although Interstate 80 was suspected as a source because fast-moving vehicles may re-entrain and distribute dust on the roadway, it did not show elevated concentration of $PM_{10-2.5}$.

ANOVA showed that temporal variation dominated the total variability in $PM_{10-2.5}$ but that spatial variability was still important. $PM_{10-2.5}$ was consistently lower throughout the sampling area during Week 3 when substantial rainfall occurred (Fig. 3). Rain may have directly removed coarse particles from the air by impaction and reduced the generation of coarse particles because surfaces of common sources were wet. This phenomenon dominated the between-week versus within-week analysis, which showed that between-week (or temporal) variability accounted for 70% of the total variability. Despite this dominating rain event, within-week variability (spatial plus random error) still accounted for 30% of the total variability.

The lack of correlation between semivariance and distance between sites (Fig. 4) demonstrates that $PM_{10-2.5}$ measured with passive samplers were spatially independent. Such independence is required to apply many statistical analyses of pollutant spatial distribution. For example, spatially correlated sites would be inappropriately weighted greater than other sites in the calculation of mean $PM_{10-2.5}$. Spatial independence also simplifies statistical models used to predict $PM_{10-2.5}$ because the model does not have to account for the autocorrelation. Moreover, spatial independence is economical because it avoids redundancy in sampling.

These results confirm the validity and effectiveness of siting passive samplers with data from mobile sampling for $PM_{10-2.5}$. Site location and number are critically important for effectively capturing intra-city variability of air pollutants. However, effective siting requires preliminary knowledge of the temporal and spatial variability of the pollutant in the measurement area, referred to as a demand surface by Kanaroglou et al. (2005). In this work, a preliminary demand surface was constructed with data from a real-time instrument that measured $PM_{10-2.5}$ throughout the city during a 4-day period. This surface was used first to determine the number of sites (i.e. 33) needed to capture about 95% of the total variability with 95% confidence interval. Then, these sites were located such that spatial autocorrelation was minimized.

The primary limitation of this work was that only one city was investigated during a short period of time. Thus, the findings of this study may not necessary hold for other cities and seasons. Further research comparing different seasons and across different cities is warranted to generalize the robustness of passive sampling to capture spatial variability not only in $PM_{10-2.5}$ but also in other air pollutants.

In conclusion, this work used passive samplers sited with data from mobile sampling to show that $PM_{10-2.5}$ is heterogeneous at a local scale in a medium-sized Midwest US City. These results may aid us in quantifying the degree of exposure misclassification in epidemiological studies that rely on data from a central monitoring station. This work also confirms the validity and effectiveness of siting passive samplers with data from mobile sampling for $PM_{10-2.5}$. This approach aimed to maximize variance and minimize spatial autocorrelation enabled data obtained through passive sampling to be spatially independent. The inexpensive and simple nature of these methods when integrated into compliance networks could be used to assess exposure to ambient $PM_{10-2.5}$ at a local scale, such as a household location. In turn, these estimates may be incorporated into epidemiological studies to more effectively investigate associations between adverse health outcomes and exposure to coarse particulate matter.

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