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Urban-scale Spatial-temporal Variability of Black Carbon and Winter Residential Wood Combustion Particles

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ABSTRACT

It has been suggested that certain organic aerosol components of wood smoke have enhanced ultraviolet absorption at 370 nm relative to 880 nm in two-wavelength aethalometer black carbon (BC) measurements. This enhanced absorption could serve as an indicator of wood combustion particles ("Delta-C" = $UVBC_{370nm}$ – BC_{880nm}). From August 2009 to October 2010, week-long mobile monitoring campaigns were conducted during each season in Rochester, New York. The temporal and spatial variations of BC and Delta-C were investigated at twelve monitoring sites. A portable two-wavelength aethalometer housed in Clarkson's Mobile Air Pollution Lab was used for data collection. The average BC concentrations were 0.94 μg/m^3, 0.68 μg/m^3, 0.47 μg/m^3, and 0.81 μg/m^3 in summer, winter, spring, and fall, respectively. BC and Delta-C hotspots were identified. Coefficients of divergence (COD) and correlation coefficients (r) were calculated between site pairs to assess the spatial and temporal heterogeneity. High spatial divergence but uniform temporal variation in BC were found for these sites. Winter residential wood combustion (RWC) particles exhibited high spatial heterogeneity as well. In epidemiological studies, BC particles data from a central monitoring site are generally used as the basis in population exposure estimation. These results suggest that one central monitoring site may not adequately represent the actual BC and RWC particle exposure over a whole urban area.

Keywords: Spatial-temporal variability; Black carbon (BC); Delta-C; Residential wood combustion (RWC); Aethalometer; Coefficient of divergence.

INTRODUCTION

Black carbon (BC) is generally considered to be a traffic-related airborne particle produced as a combustion by-product. Typically more than 90% of BC is present in the $PM_{2.5}$ (particles with an aerodynamic diameter less than or equal to 2.5 μm) size fraction (Viidanoja *et al*., 2002). BC emitted from traffic sources has been associated with increased risk of asthma and bronchitis in children (Kim *et al*., 2004). In a cohort study conducted in East Boston, Massachusetts, exposure to traffic-related BC independently predicted decreased lung function in urban women (Suglia *et al*., 2008). Positive associations between blood pressure and BC were observed in elderly men (Mordukhovich *et al*., 2009). BC is also associated with increased risk of emergency myocardial infarction hospitalization (Zanobetti and Schwartz, 2006).

Assessing human exposure to BC requires evaluation of its spatial variability in different urban scales. Watson and Chow (2001) concluded that a carefully selected fixed-site monitor can reasonably represent exposures in its surrounding neighborhood even if many local sources affect the monitor. However, a number of recent studies indicate spatial heterogeneity in ambient BC concentrations. In New York City, BC concentrations were concurrently measured at two urban sites 9.1 km apart using two aethalometers in winter 2004. Moderate spatial heterogeneity was observed in the BC concentrations (Venkatachari *et al*., 2006). In Long Beach, California, high spatial variability was observed in the < 0.25 μm, 0.25–2.5 μm and > 2.5 μm PM fractions for BC in the winter of 2005, based on data collected concurrently at four sites within four miles of one another (Krudysz *et al*., 2008). In Detroit, Michigan and Cleveland, Ohio, the site-to-site variability in BC concentrations was significant at both neighborhood (0.5–4 km) and urban (4–100 km) scales (Snyder *et al*., 2010).

Comprehensive BC exposure studies have been conducted in the greater Boston area. Gryparis *et al*. (2007) propose latent variable semiparametric regression models for modeling the spatial and temporal variability of BC and elemental carbon (EC). Brown *et al*. (2008) found that ambient EC concentrations were weakly associated with corresponding

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personal exposure level for 25 individuals. Personal exposures to EC were higher for those individuals living close to a major road (Brown *et al*., 2009). Thus, intraurban variation of BC needs to be well understood in human exposure and epidemiological studies.

In the United States, residential wood combustion (RWC) is responsible for 6.9% of the national primary $PM_{2.5}$ emissions (U.S. EPA, 2006). These emissions are larger than the contribution of on-road (2.5%) and similar to that for off-road (7.3%) mobile sources. Source apportionment studies suggest that the RWC is the largest contributor to $PM_{2.5}$ in rural areas of New York State (NYSERDA, 2008), and it also accounts for 15% of $PM_{2.5}$ in the southeastern Unites States (Zheng *et al*., 2002). Wood combustion emits significant quantities of air pollutants and known carcinogens, such as benzene and polycyclic aromatic hydrocarbons (PAHs). Exposure of healthy subjects to wood smoke caused an increase in antioxidant airway responses (Sehlstedt *et al*., 2010). Naeher *et al*. (2007) reviewed the published results regarding wood smoke health effects. The summary shows that significant exposures to ambient wood smoke do occur worldwide and it could cause acute and chronic illness. In addition, spatial heterogeneity in wood smoke was observed among six sites in a residential neighborhood in northern Sweden (400 m × 400 m) during the winter 2006 (Krecl *et al*., 2010). Therefore, a central monitoring site may not adequately assess exposure to RWC particles either.

The present study measured the spatial-temporal variability of BC concentrations over four seasons and the spatial variability of winter RWC particles in Rochester, NY, using aethalometer data collected at twelve locations. The heterogeneity of the BC and RWC particle concentrations within the sampling area was explored. The feasibility of using highly time-resolved instruments installed in mobile monitoring labs to identify the hot spots of particles over a medium-size urban area was also studied.

MATERIAL AND METHODS

Sampling Sites

The 2008 population of Rochester was 206 886, the third largest city in New York. Aethalometer measurements were made at twelve monitoring sites in East Rochester, New York from August 2009 to October 2010. Hourly $PM_{2.5}$ mass, SO_2, O_3, CO, ultrafine particle (UFP) number concentrations, wind speed and wind direction, ambient temperature and relative humidity were measured by the New York State Department of Environmental Conservation (NYS DEC) at the monitoring site. Geographical data for the monitoring sites, pollutant data measured at the DEC site, and meteorological conditions during the sampling period are summarized by Wang *et al*. (submitted). Fig. 1 shows the locations of the monitoring sites and the major roadways distributed in an approximate 9 km × 9 km area. Four one-week sampling campaigns were conducted in August 12–18, 2009 (summer), December 13–18, 2009 (winter), April 23–28, 2010 (spring) and October 18–23, 2010 (fall). Aethalometer measurements were made over 12 minutes at each site on each day during two separate periods, 7:00–10:00 and 16:00–19:00, during which the total traffic volume on the nearby highways was relatively stable (see Fig. 2). There was relatively constant traffic volumes over these three years.

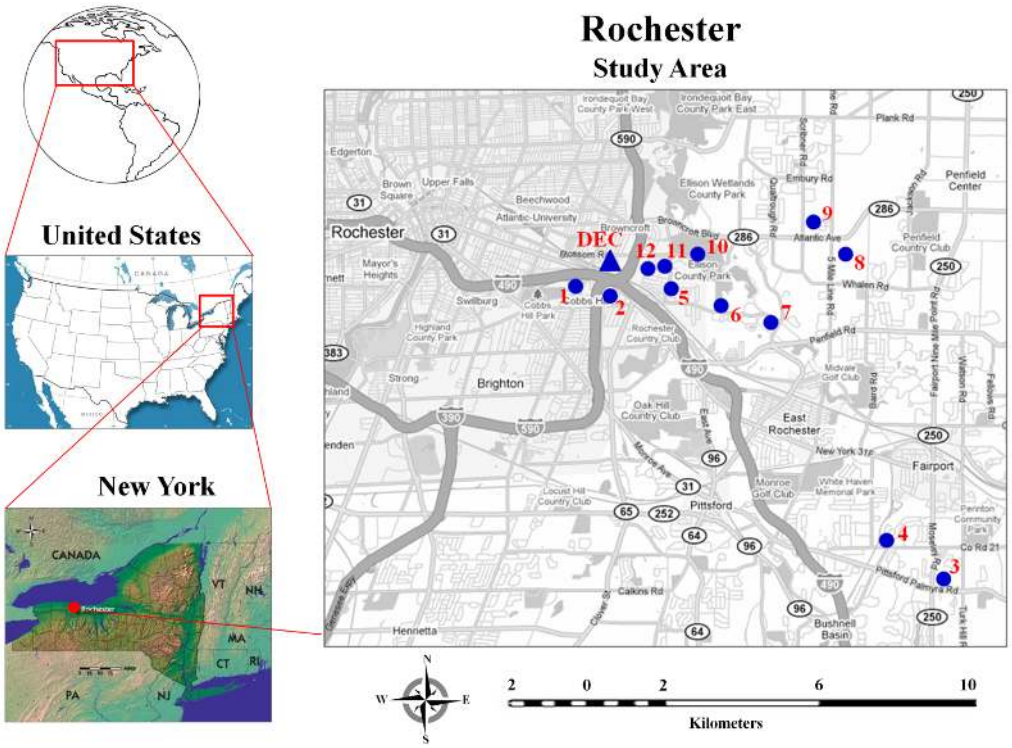

Fig. 1. Location of the monitoring sites and major roadways in the study area.

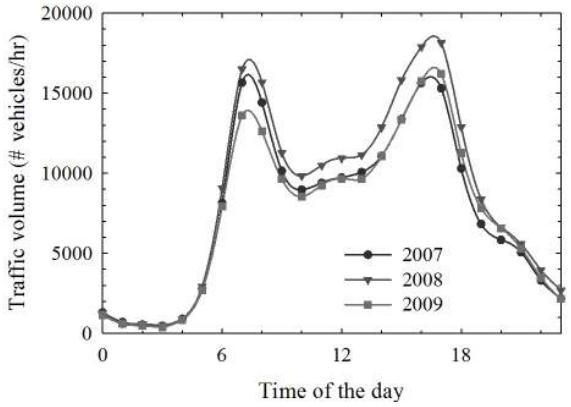

Fig. 2. Diurnal variations of total traffic volume on both I-490 and I-590 measured near the DEC site on weekdays in 2007, 2008, and 2009 (Monitoring dates: Nov 13–16, 2007; Oct 7–10 and 13–14, 2008; Aug 24–27, 2009).

Instrumentation

Measurements were conducted with a two-wavelength aethalometer (model AE-42, Magee Scientific, Berkeley, CA) at a time resolution of 5 min housed in Clarkson's Mobile Air Pollution Lab (MAPL) (Ogulei *et al*., 2007). The two-wavelength aethalometer measures the optical absorption of ambient PM at 880 nm (BC) and 370 nm (UVBC) (Hansen *et al*., 1984). Certain organic aerosol components of wood combustion particles have enhanced optical absorption at 370 nm relative to 880 nm (Jeong *et al*., 2004; Park *et al*., 2006). A calculated variable, Delta-C signal ($UVBC_{370nm} - BC_{880nm}$), has been suggested to serve as an indicator of wood combustion particles, but is not a direct quantitative measurement of mass concentrations (Allen *et al*., 2004; Wang *et al*., 2010, 2011).

Data Analysis

The aethalometer estimates the BC mass concentrations from the rate of change of light transmission through a filter. As the filter gets darker, the measured BC concentration is underestimated. In this study, the loading effect of aethalometer data was corrected using Eq. (1)

$$BC_{corrected} = (1 + K \cdot ATN) \cdot BC_{non\text{-}corrected} \quad (1)$$

where ATN is the attenuation, K is the correcting factor (Virkkula *et al*., 2007). The K_i factor was then used to correct all of the two-wavelength (370 nm and 880 nm) BC data obtained for filter spot i in Eq. (1). This algorithm has been used in other recent studies (Park *et al*., 2010; Wang *et al*., 2011). The average 880 nm-wavelength K factors were 0.0068, 0.0017, 0.0062, and 0.0081 for spring, summer, fall, and winter, respectively. The average 370 nm-wavelength K factors were 0.0054, 0.0042, 0.0054, and 0.0058 for spring, summer, fall, and winter, respectively.

Wilson *et al*. (2005) have suggested that absolute and relative measures of homogeneity based on measurements at several monitoring sites and sufficiently large sample sizes (> 50 samples) may reduce possible misclassification. Pearson correlation coefficients were used to explore the temporal relationships among the sampling sites. Relative spatial variability was assessed by calculating the coefficients of divergence (COD). The COD is defined as

$$COD_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}[(x_{ij} - x_{ik})/(x_{ij} + x_{ik})]^2} \quad (2)$$

where x_{ij} is the i^{th} averaged concentration for a given pollutant measured at site j, j and k are two different sites, and p is the number of observations (Wongphatarakul *et al*, 1998; Pinto *et al*., 2004).

A COD value equal to zero means the concentrations are identical at both sites, while a value approaching one indicates substantial heterogeneity. COD values greater than approximately 0.20 indicate relatively heterogeneous spatial distributions (Pinto *et al*., 2004; Wilson *et al*, 2005). COD analysis has been used to evaluate the spatial variability of carbonaceous particles (Venkatachari *et al*., 2006; Krudysz *et al*., 2008; Krecl *et al*., 2010; Sarnat *et al*., 2010). It complements the Pearson correlation coefficients (Moore *et al*, 2009).

RESULTS AND DISCUSSION

Seasonal and Spatial Variations of Black Carbon Concentrations

Fig. 3 shows the BC concentrations measured at the twelve sites during the morning (AM) and afternoon (PM) campaigns in each season. The values shown in Fig. 3 are also given in Table 1 (mean ± standard deviation). The average BC concentrations were 0.94 μg/m^3, 0.68 μg/m^3, 0.47 μg/m^3, and 0.81 μg/m^3 in summer, winter, spring, and fall, respectively. The overall AM average BC concentration was 0.92 μg/m^3, approximately 70% greater than the PM value (0.54 μg/m^3). Lower atmospheric mixing height and ambient temperature in the morning compared to the afternoon are likely to be responsible for this concentration difference.

Among the summer AM samples (Fig. 3(a)), the highest and lowest BC concentrations were observed at sites 6 and 4, respectively. The second highest BC concentration was at site 9, where a number of heavy-duty diesel-powered construction vehicles were being operated during the sampling period. Visible soot particles were emitted from the tailpipe of these vehicles. In the afternoon (Fig. 3(b)), the highest and lowest BC concentrations were found at sites 12 and 5, respectively. Although these two sites are only 1 km apart, their BC concentrations were statistically different (student t-test, $p < 0.01$). For both the AM and PM campaigns, BC concentrations at site 12 were 17% higher compared to site 11 (1.03 μg/m^3 vs. 0.88 μg/m^3), possibly as the result of distances to the nearby highway (50 m for site 12 and 300 m for site 11).

On winter mornings, BC concentrations were the highest and the lowest at sites 12 and 10, respectively (Fig. 3(c)). Site 6 became a hotspot for BC during winter afternoons (Fig. 3(d)). Since there are no nearby highways, other activities such as the RWC and off-road diesel engines must be the major BC sources. In the spring, sites 5 and 8

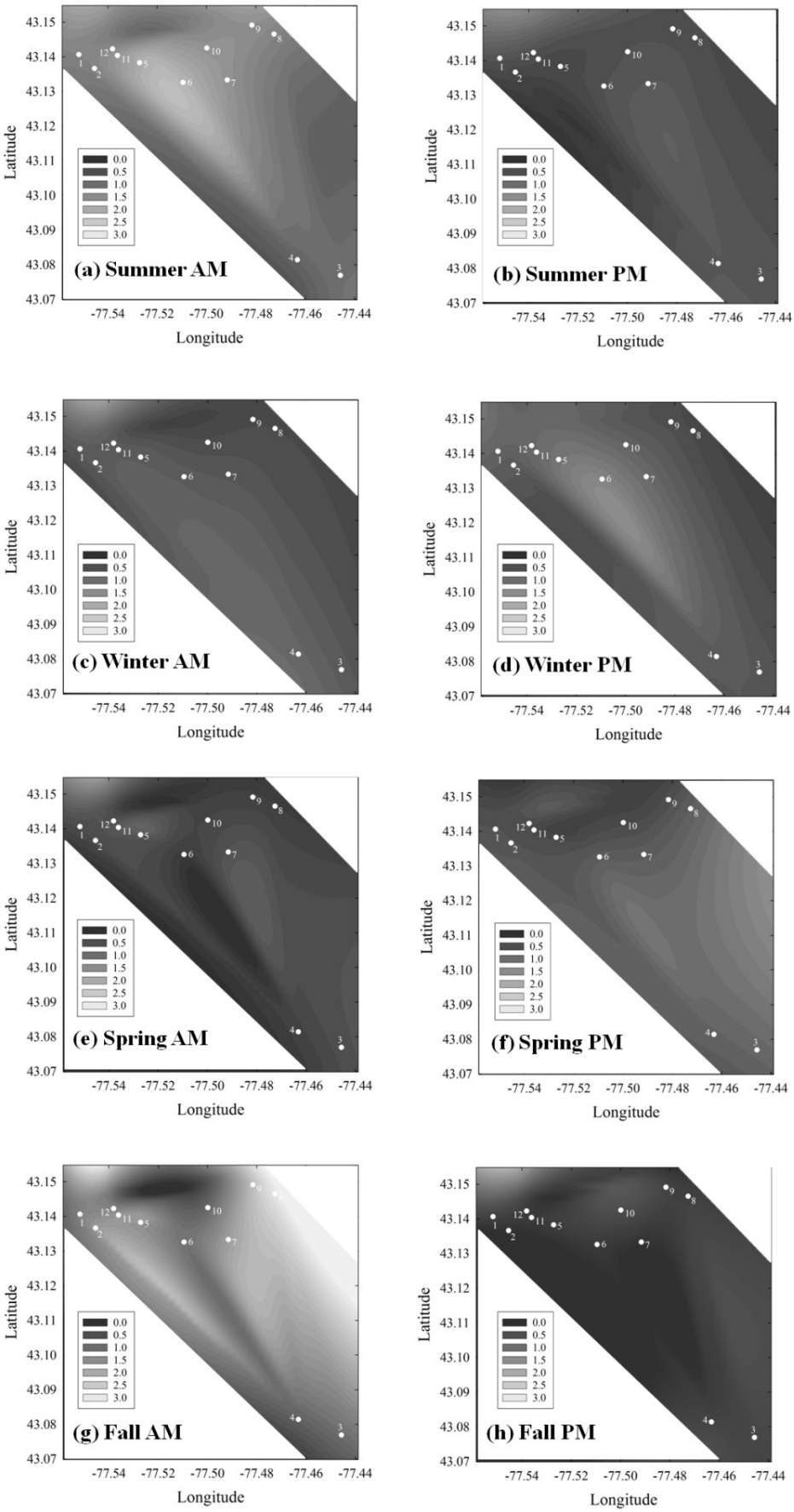

Fig. 3. Contour plots of the BC concentrations.

Table 1. Summary of BC concentrations (μg/m^3). Numbers in the parentheses represent one standard deviation.

Site	Summer AM	Summer PM	Winter AM	Winter PM	Spring AM	Spring PM	Fall AM	Fall PM
1	1.33 (0.12)	0.67 (0.23)	0.59 (0.07)	0.92 (0.43)	0.44 (0.05)	0.48 (0.12)	0.90 (0.17)	0.48 (0.35)
2	1.31 (0.18)	0.47 (0.17)	0.65 (0.19)	0.72 (0.16)	0.43 (0.04)	0.50 (0.20)	1.42 (0.32)	0.19 (0.09)
3	1.07 (0.33)	0.57 (0.13)	0.62 (0.18)	0.55 (0.21)	0.49 (0.18)	0.78 (0.42)	0.81 (0.68)	0.32 (0.15)
4	0.83 (0.34)	0.60 (0.21)	0.81 (0.02)	0.64 (0.22)	0.29 (0.12)	0.77 (0.73)	0.74 (0.10)	0.33 (0.18)
5	1.66 (0.42)	0.40 (0.17)	0.56 (0.16)	1.00 (0.50)	0.65 (0.08)	0.32 (0.11)	1.25 (0.24)	0.15 (0.03)
6	2.08 (0.88)	0.44 (0.28)	0.63 (0.16)	1.23 (0.43)	0.32 (0.05)	0.50 (0.41)	1.10 (0.22)	0.24 (0.05)
7	1.62 (0.36)	0.60 (0.28)	0.59 (0.11)	0.96 (0.33)	0.49 (0.02)	0.67 (0.33)	1.62 (0.87)	0.05 (0.02)
8	1.42 (0.48)	0.42 (0.37)	0.39 (0.07)	0.36 (0.10)	0.16 (0.07)	0.64 (0.36)	3.04 (0.19)	0.20 (0.03)
9	1.77 (0.64)	0.49 (0.40)	0.38 (0.11)	0.46 (0.12)	0.18 (0.03)	0.26 (0.20)	2.57 (0.23)	0.09 (0.06)
10	1.36 (0.65)	0.48 (0.16)	0.30 (0.09)	0.62 (0.42)	0.21 (0.07)	0.23 (0.15)	0.48 (0.44)	0.62 (0.57)
11	1.05 (0.20)	0.70 (0.29)	0.64 (0.23)	0.88 (0.43)	0.27 (0.16)	0.60 (0.47)	0.70 (0.45)	0.29 (0.04)
12	1.22 (0.28)	0.83 (0.46)	0.87 (0.44)	0.85 (0.48)	0.56 (0.15)	0.49 (0.33)	1.22 (0.34)	0.63 (0.22)

represented the highest and lowest BC concentrations in the morning over the sampling area (Fig. 3(e)). During the afternoon, the highest BC values were at sites 3 and 4. Site 10 seemed to have the minimum BC concentration (Fig. 3(f)). The AM average BC concentrations were lower than the PM values for both winter and spring.

During fall mornings (Fig. 3(g)), the maximum and minimum BC concentrations were observed at sites 8 and 10, respectively. The BC concentration at site 8 was also the highest during the entire four-season sampling period, although it was located 4500 m from the major highways. It seemed that non-highway BC sources must be significantly affecting the local air quality under favorable weather conditions. Site 12 and site 7 represented the highest and lowest BC concentrations in the afternoon, respectively (Fig. 3(h)). Similar to the summer campaign, the BC concentration at site 12 was statistically higher compared to site 11 during the fall (student t-test, $p < 0.01$). People living at the twelve sites located in the 9 km × 9 km urban area exposed to different BC concentrations at different time of the day in different seasons.

Spatial Variability in Black Carbon Concentrations

COD Values

COD values were calculated for all site pairs during four seasons and are given in Table 2. Measurements at sites 3 and 4 and the other ten sites were made on different days in the summer, so the COD values between sites 3 and 4 and the other sites are not included in the table. COD values larger than 0.2 indicate spatial heterogeneity, and are highlighted in colors between yellow (lowest) and red (highest). Most of the site pairs displayed moderate to high spatial heterogeneity. Four site pairs (sites 1–2, sites 3–4, sites 8–9, and sites 11–12) were selected because the two sites in each pair are spatially close and there was a short time lag between the measurements at the two sites (~5 min).

Sites 1 and 2 are physically close (840 m) and near to the I-490/I-590 intersection (~250 m). COD values between the two sites show spatial homogeneity during summer, winter, and spring. Relatively high divergence was observed in the fall indicating moderate spatial heterogeneity between the two sites. Sites 3 and 4 were separated by 1900 m and showed differences in BC concentrations. Moderate divergence (COD > 0.24) was seen in the BC concentrations during the spring and fall. Site 3 was 100 m from a busy local road while site 4 was a background site separated from the major highways. Traffic on local streets near site 4 made significant contributions to BC concentrations at this location.

Sites 8 and 9 (1000 m apart) show the spatial heterogeneity in winter, spring, and fall. The COD value of 0.17 between sites 8 and 9 during summer indicates the off-road diesel engines operated likely impacted the air quality as far as 1000 m from the sources. Sites 11 and 12 were located 300 m and 50 m from I-590, respectively. There were no other major BC sources near these sites. Moderate heterogeneity between the two sites was found in summer, spring, and fall. Low ambient temperature and different predominant wind direction (southeast) during the winter may weaken the spatial heterogeneity between the two sites.

The COD has been used in other locations to characterize the spatial homogeneity of BC concentrations. Krudysz *et al*. (2008) measured intra-community spatial variation of size-fractionated BC concurrently at four sites within four miles of one another in the Long Beach, CA area. High spatial variability (COD > 0.4) was observed in the 0.25–2.5 μm and > 2.5 μm PM fractions. Venkatachari *et al*. (2006) measured BC at two sites (9.1 m apart) in New York City during the winter of 2004. The COD value for BC was 0.29 indicating some degree of spatial heterogeneity in the ambient BC concentrations. Our results are comparable to these findings suggesting that one central monitoring site may be not adequate to estimate BC exposures over an urban area.

Correlation Coefficients

In addition to the COD spatial variability analysis, Pearson correlation coefficients (r) were used to assess the temporal association between two sampling sites. Table 3 summarizes the Pearson correlation coefficients for all the same site pairs shown in Table 2. Negative values indicating inverse correlations are shown with white background. All the positive values are highlighted with colors between yellow (lowest value = 0.0) and red (highest value = 1.0). Moderately strong positive correlations exist widely for all site pairs during summer. In the fall, BC concentrations

Table 2. Results of Coefficient of Divergence (COD) analysis for BC.

site	2	3	4	5	6	7	8	9	10	11	12
1	0.16	--	--	0.26	0.31	0.17	0.36	0.31	0.19	0.20	0.20
2		--	--	0.17	0.29	0.19	0.33	0.29	0.15	0.21	0.25
3			0.11	--	--	--	--	--	--	--	--
4				--	--	--	--	--	--	--	--
5					0.23	0.23	0.31	0.26	0.21	0.35	0.32
6						0.25	0.23	0.18	0.20	0.37	0.36
7							0.29	0.23	0.19	0.24	0.17
8								0.17	0.29	0.34	0.35
9									0.26	0.34	0.34
10										0.23	0.24
11											0.21

summer

site	2	3	4	5	6	7	8	9	10	11	12
1	0.16	0.31	0.28	0.14	0.17	0.13	0.37	0.32	0.38	0.29	0.30
2		0.23	0.20	0.20	0.21	0.16	0.35	0.30	0.36	0.22	0.19
3			0.19	0.35	0.39	0.35	0.23	0.17	0.20	0.28	0.28
4				0.30	0.28	0.25	0.36	0.24	0.32	0.19	0.20
5					0.16	0.19	0.42	0.34	0.37	0.30	0.27
6						0.08	0.44	0.38	0.44	0.27	0.25
7							0.40	0.33	0.41	0.23	0.25
8								0.21	0.28	0.39	0.46
9									0.21	0.33	0.16
10										0.33	0.37
11											0.16

winter

site	2	3	4	5	6	7	8	9	10	11	12
1	0.09	0.21	0.37	0.23	0.25	0.15	0.36	0.37	0.31	0.36	0.31
2		0.26	0.32	0.23	0.22	0.14	0.38	0.42	0.47	0.38	0.35
3			0.29	0.34	0.32	0.18	0.32	0.49	0.53	0.31	0.29
4				0.40	0.28	0.30	0.31	0.42	0.53	0.26	0.36
5					0.27	0.30	0.45	0.40	0.47	0.41	0.28
6						0.25	0.30	0.30	0.45	0.36	0.41
7							0.37	0.49	0.52	0.40	0.36
8								0.38	0.46	0.24	0.39
9									0.30	0.41	0.47
10										0.47	0.51
11											0.27

spring

site	2	3	4	5	6	7	8	9	10	11	12
1	0.32	0.25	0.15	0.37	0.25	0.58	0.48	0.58	0.37	0.31	0.23
2		0.34	0.29	0.14	0.18	0.43	0.28	0.33	0.53	0.37	0.41
3			0.24	0.40	0.32	0.59	0.48	0.58	0.48	0.38	0.40
4				0.31	0.19	0.57	0.47	0.55	0.37	0.21	0.30
5					0.17	0.43	0.31	0.33	0.54	0.34	0.43
6						0.52	0.34	0.45	0.47	0.23	0.31
7							0.51	0.36	0.70	0.62	0.64
8								0.30	0.63	0.48	0.48
9									0.70	0.58	0.59
10										0.34	0.43
11											0.35

fall

Table 3. Results of Pearson correlation coefficient (r) analysis for BC.

site	2	3	4	5	6	7	8	9	10	11	12
1	0.88	--	--	0.85	0.83	0.88	0.74	0.87	0.67	0.55	0.43
2		--	--	0.93	0.87	0.84	0.81	0.79	0.88	0.71	0.52
3			0.90	--	--	--	--	--	--	--	--
4				--	--	--	--	--	--	--	--
5					0.92	0.84	0.88	0.91	0.96	0.61	0.59
6						0.91	0.86	0.90	1.00	0.75	0.68
7							0.81	0.90	0.69	0.63	0.68
8								0.89	0.94	0.66	0.84
9									0.79	0.55	0.63
10										0.62	0.66
11											0.57

summer

site	2	3	4	5	6	7	8	9	10	11	12
1	0.47	-0.44	-0.93	0.88	0.62	0.68	0.12	-0.34	-0.01	-0.40	-0.34
2		-0.30	-0.47	0.63	0.46	0.54	-0.29	-0.37	0.26	0.26	0.57
3			0.09	-0.17	-0.90	-0.88	0.70	0.24	0.80	0.05	-0.10
4				-0.93	-0.27	-0.37	-0.60	0.73	-0.10	0.59	0.58
5					0.65	0.64	0.05	-0.12	0.39	-0.08	0.01
6						0.98	-0.63	0.31	-0.04	0.32	0.19
7							0.94	0.98	0.87	0.90	0.92
8								-0.51	0.18	-0.66	-0.72
9									0.43	0.66	0.27
10										0.48	0.85
11											0.85

winter

site	2	3	4	5	6	7	8	9	10	11	12
1	0.80	1.00	0.80	0.23	0.88	0.90	0.88	0.93	0.47	0.89	0.39
2		0.70	0.95	0.19	0.94	0.94	0.41	0.86	0.01	0.74	0.00
3			0.79	0.03	0.66	0.79	0.89	0.58	-0.12	0.86	0.57
4				0.01	0.91	0.96	0.61	0.81	-0.10	0.90	0.11
5					0.24	0.09	-0.35	0.14	-0.05	-0.15	0.25
6						0.98	0.35	0.98	0.24	0.00	-0.19
7							0.56	0.92	0.15	0.53	0.02
8								0.30	-0.22	0.86	0.59
9									0.49	-0.38	-0.29
10										-0.67	-0.64
11											0.91

spring

site	2	3	4	5	6	7	8	9	10	11	12
1	0.83	0.76	0.92	0.68	0.68	0.78	0.76	0.77	0.16	0.32	0.66
2		0.77	0.91	0.89	0.88	0.96	0.98	0.99	-0.28	0.47	0.70
3			0.59	0.40	0.38	0.89	0.63	0.65	-0.45	-0.19	0.16
4				0.91	0.91	0.79	0.92	0.92	0.15	0.65	0.89
5					1.00	0.75	0.96	0.95	-0.03	0.81	0.92
6						0.73	0.95	0.94	0.01	0.83	0.93
7							0.90	0.91	-0.46	0.23	0.49
8								1.00	-0.22	0.62	0.80
9									-0.22	0.60	0.79
10										0.37	0.37
11											0.92

fall

measured at site 10 did not seem to vary with temporal patterns similar to the other sites. Relatively overall low r values for summer and spring suggest weak temporal uniformity.

In the fall, a group of negative r values were found between site 10 and the other sites indicating temporal non-uniformity. The COD value was greater than 0.48 during the same period suggesting high spatial heterogeneity. During winter, 40% of the r values were smaller than zero corresponding to that 22% of the COD values during the same period that were smaller than 0.20. These values are indicative of similar conclusions in the temporal and spatial divergence. Lianou *et al.* (2007) reported that high r values between paired sites would only imply uniform temporal variation. Both r and COD values are required to evaluate the spatial-temporal variability in ambient pollutant concentrations.

Spatial Variation of Winter Wood Combustion Particles

Delta-C values were calculated for all four seasons to

qualitatively indicate the concentration of wood combustion particles in this study. The spatial variations of Delta-C values in summer, spring, and fall are not included because of the large number of negative values indicating no impacts from wood smoke (Wang *et al*., 2011). In addition, negative values are not suitable for the COD/correlation analysis. Fig. 4 shows the spatial variations of Delta-C values during the AM and PM campaigns in winter. The average PM Delta-C value was 0.30 μg/m^3, 50% greater compared to the AM value.

The highest and lowest Delta-C values during the AM were 0.32 μg/m^3 and 0.02 μg/m^3 observed at site 2 and site 12, respectively (Fig. 4(a)). These two sites were both near the intersection of I-490 and I-590 highways and only 1.2 km apart. The completely different Delta-C levels at the two close sites suggest that Delta-C is a reasonable indicator of wood combustion particles in the presence of traffic-related particle sources. In the winter PM observations (Fig. 4(b)), sites 5 and 3 represent the highest (0.67 μg/m^3) and lowest (0.08 μg/m^3) Delta-C levels in the study area, respectively. The RWC activities in the school at site 5 during the sampling period played an important role in the ambient Delta-C levels (Wang *et al*., 2011), making people living near the site 5 have the highest wood smoke exposure. Fig. 4(b) also shows that winter RWC is a local source of particle pollution.

Spatial Variability in Winter Wood Combustion Particles

COD Calues

COD values for Delta-C values calculated for all site pairs in the winter are presented in Table 4. All the values are greater than 0.2 indicating moderate spatial heterogeneity in winter residential wood smoke across the study area. Larger COD values (min: 0.43, max: 0.92) between sites 3 and 4 and the other ten sites indicated higher spatial non-uniformity of Delta-C values between the southeastern part and the northern part of the sampling area. As mentioned previously, site 5 was expected to be impacted by the nearby RWC activities. The Delta-C values measured at site 5 were the highest suggesting the largest wood smoke contribution. The COD values between site 5 and the other nearby sites (sites 2, 6, 10, 11 and 12) are between 0.34 (sites 5 and 6) and 0.76 (sites 5 and 12). The results imply that winter RWC smoke is a local pollutant and it could significantly affect local air quality within a short distance from the sources.

Correlation Coefficients

Table 5 summarizes the intersite Pearson correlation coefficients for winter Delta-C values. Most r values are greater than 0.70 indicating temporal uniformity of winter wood smoke across the area. However, a number of negative r values existing between site 5 and the other sites show significant temporal heterogeneity. Combing with the COD results, local RWC activities could influence the temporal-spatial variability of wood smoke exposure at the urban scale.

CONCLUSIONS

High spatial heterogeneity and moderate temporal homogeneity in BC concentrations were observed across the study area. Winter RWC particles exhibited high spatial heterogeneity as well. Their local sources played an important role in the exposure level of people living in the nearby communities. The results indicate that one central monitoring site may not provide an accurate approximation of the actual human exposure to BC and RWC particles over the whole urban area. In epidemiological studies assessing adverse health effects from long-term exposure, multiple monitoring sites covering the spatial variability will be required to obtain accurate exposure/risk coefficients. Using highly time-resolved instruments installed in mobile monitoring labs proved to be a useful approach to map the spatial-temporal variability of particles over a medium-size urban area.

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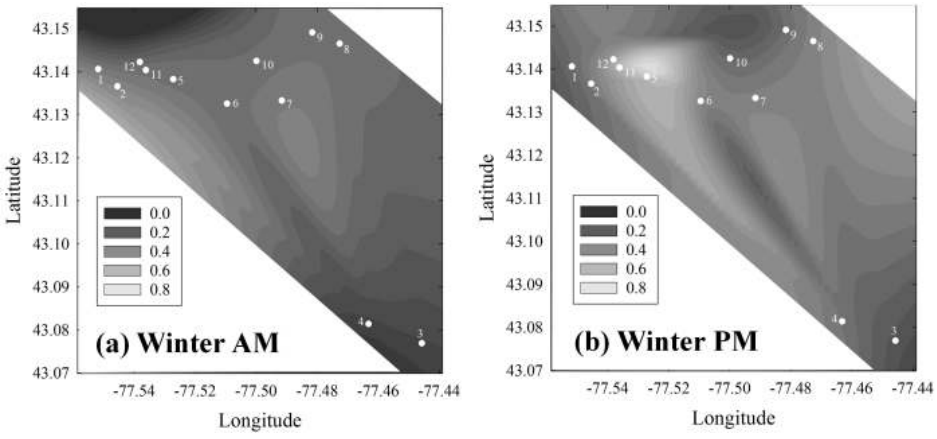

Fig. 4. Contour plots of the winter Delta-C values.

Table 4. Results of COD analysis for winter Delta-C values.

site	2	3	4	5	6	7	8	9	10	11	12
1	0.31	0.45	0.75	0.39	0.35	0.53	0.44	0.45	0.65	0.78	0.81
2		0.54	0.67	0.41	0.25	0.52	0.24	0.39	0.63	0.71	0.73
3			0.66	0.68	0.67	0.66	0.53	0.43	0.80	0.92	0.84
4				0.72	0.70	0.59	0.66	0.68	0.74	0.82	0.61
5					0.34	0.48	0.51	0.45	0.61	0.73	0.76
6						0.55	0.38	0.51	0.65	0.77	0.79
7							0.52	0.43	0.50	0.68	0.73
8								0.38	0.62	0.72	0.79
9									0.59	0.69	0.78
10										0.69	0.63
11											0.78

Table 5. Results of r analysis for winter Delta-C values.

site	2	3	4	5	6	7	8	9	10	11	12
1	0.59	0.19	-0.34	0.16	0.24	0.18	-0.20	0.31	0.01	-0.17	-0.3(
2		0.85	0.94	0.02	0.82	0.87	0.65	0.90	0.73	0.63	0.9
3			0.79	-0.38	0.80	0.85	0.82	0.84	0.56	0.79	0.7
4				0.01	0.92	0.93	0.89	0.85	0.74	0.89	0.9
5					0.29	-0.04	-0.16	-0.02	-0.09	0.02	-0.01
6						0.84	0.70	0.79	0.59	0.76	0.8
7							0.94	0.98	0.87	0.90	0.9
8								0.90	0.87	0.98	0.9
9									0.90	0.86	0.8
10										0.85	0.9
11											0.9

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