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Characterization of Ultrafine Particles and Other Traffic Related Pollutants near Roadways in Beijing

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ABSTRACT

Developing countries, such as China, are facing serious air pollution issues due to fast economic development. In this study, traffic related air pollutants, including number concentration of ultrafine particles (UFPs, diameter < 100 nm), mass concentrations of $PM_{2.5}$ and black carbon (BC) were measured near the Peking University (PKU) campus in Beijing in December 2011. Data were collected concurrently at a roadway site and on PKU campus. Meteorological data were collected at approximately 40 meters northeast from the roadway sampling site. The traffic density was determined from recorded video footage. Roadside UFP and $PM_{2.5}$ concentrations were not significantly higher than on campus. A statistically significant Pearson's correlation of 0.75 was found between BC and $PM_{2.5}$ mass concentrations. No apparent correlation was found between wind speed and UFP number concentrations, but strong log-decay correlations were found between wind speed and $PM_{2.5}$ ($PR_{2.5}$ mass concentrations and UFP number concentrations were higher at the campus site than at the roadway site. This suggests there were potential local emission sources on campus. Temporal profile of UFPs at the campus site peaked around lunch and dinner time, suggesting emissions from the surrounding restaurants and cafeteria that used Chinese-style cooking might have contributed to the observed $PM_{2.5}$ and UFP levels on campus.

Keyword: Ultrafine particles; PM_{2.5}; Black carbon; Beijing; Roadway; Cooking emissions.

INTRODUCTION

Air pollution studies conducted in the US and Europe have shown a consistent relationship between increases in particulate matter (PM) exposure and increases in human mortality and morbidity. In developing countries, such as China, the rapid industrialization and urbanization have led to huge increases of PM emissions. Beijing is one of the megacities in China with a population of 20.69 million. According to 2012 Beijing Environmental Statement, the 2012 annual average PM_{10} (particles with aerodynamic diameter $\leq 10~\mu m$) mass concentration was $109~\mu g/m^3$, which exceeded the China Grade II National Ambient Air Quality Standards (70 $\mu g/m^3$) by 56% (Beijing Municipal

Environmental Protection Bureau, 2013).

PM pollution has been linked to adverse health outcomes, particularly respiratory and cardiovascular diseases (Schulz et al., 2005; Leitte et al., 2009; Brook et al., 2010). These diseases are prevalent in China, where air pollution levels are often orders of magnitude higher than those in developed countries (Zhang et al., 2011). Because PM deposition and related health effects depend on the particle size, $PM_{2.5}$ (fine particles with aerodynamic diameter $\leq 2.5 \mu m$) and ultrafine particles (UFP; diameter < 100 nm) have been associated more strongly with health risk than PM_{10} (Politis et al., 2008). Recent studies have shown that UFP may be particularly damaging to human health due to their ability to penetrate deeper into the lung and induce oxidative stress in deep lung tissue (Oberdorster, 2001; Donaldson et al., 2002; Politis et al., 2008).

Previous studies conducted in the United States and some European countries have demonstrated a link between vehicle traffic density and UFP, PM_{2.5}, and Black Carbon (BC) levels (Harrison *et al.*, 1997). However, a study in Beijing suggested that vehicular emissions were not the dominant factor affecting the total UFP variations in Beijing urban area (Shi *et al.*, 2007). In that study, the air sampling

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was conducted on Tsinghua University campus and no comparable data were collected near roadway to directly reflect traffic emissions. In this study, UFP and PM_{2.5} were measured concurrently at a near-road site and a campus site to evaluate the impact of traffic on UFPs in Beijing. Other emission sources that might also affect UFP variations were identified and discussed.

METHODS

Sampling Sites

Data were collected at two stationary sampling sites located on and near the Peking University (PKU) campus in Beijing, China (Fig. 1). Fig. 1(c) illustrates the orientations of the major roads and the locations of the sampling sites. The campus sampling site was located on the roof of a sixstory building on PKU main campus (Site A on Fig. 1(c)), which is about 20 meters above the ground. It was located at the east central part of PKU campus surrounded by research and office buildings, student residence halls, and dining facilities, which are all about two to six stories in height. The roadside sampling site was located at the entrance to the PKU School of Chemistry, which is outside the PKU East Gate to the main campus. This sampling site was set up at the gate guard station right outside the entrance facing Chengfu Road (Site B on Fig. 1(c)). The sampling inlets were about 1 meter above the ground. Site B was approximately 25 meters from the center of an intersection of North Zhongguancun Road and ChengfuRoad. The highest building within 300 meter distance to the sampling sites is the nine-story Founder Building located at the southeast corner of the intersection. Other buildings along the two main roads in this area are all about five to seven stories in height and are located about 20 meters from the paved pedestrian's walkways of the roads. The North

Zhongguancun Road and Chengfu Road are busy arterial roads with two to three vehicle lanes in each direction in addition to bicycle lanes and paved pedestrians walkways. Traffic on these roads includes light duty gasoline vehicles as well as diesel trucks and buses. In general, the North Zhongguancun Road runs from north to south with subway line operating underground and Chengfu Road runs from PKU east gate to east. Fig. 1 also shows the potential cooking emission sources such as restaurants and cafeterias on PKU campus. The majority of these emission sources were located approximately 200–500 meters to the east of sampling site A.

Meteorological data were collected by the PKU Automatic Meteorology Station, which is located on the third floor (~10 meter above ground) at the north building of PKU School of Physics. It is approximately 40 meters northeast from sampling site B. Data generated from this weather station were used to represent the local meteorological pattern over the sampling period.

Instrumentation and Data Collection

At Site A, a TSI water based condensation particle counter (CPC; Model 3786, TSI Inc., Shoreview, MN) was used to measure total UFP concentrations roughly from 5 nm to a few micrometers. Because in urban environments, UFPs usually constitute > 90% of total particle number concentrations, we used UFP to refer to CPC readings in this study. A TSI DustTrak photometer (Model 8520, TSI Inc., Shoreview, MN) with a 2.5 μ m inlet impactor was used to measure PM_{2.5} mass concentrations. The water based CPC was placed in an enclosed room on the roof due to its temperature sensitive nature. To prevent particle loss in the sampling line to the electrostatic charge, TSI conductive tubing was used and extended through a window to the outside ambient air. The DustTrak was placed on the roof

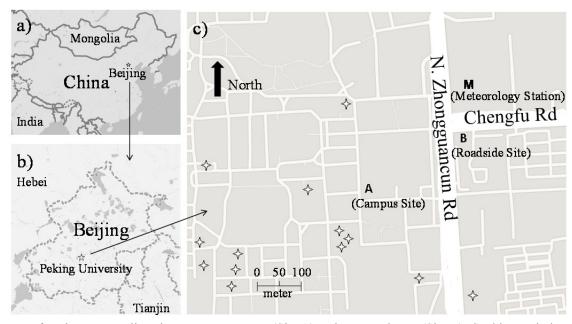


Fig. 1. Maps of stationary sampling sites on PKU campus (Site A) and near roadways (Site B). Cooking emission sources were illustrated by four-point stars on c).

directly. The reading of DustTrak was generally higher than gravimetric measurements compared with the United States Environmental Protection Agency (USEPA) designated Federal Reference Method (Chung et al., 2001; Yanosky et al., 2002). Therefore, the DustTrak data were calibrated against simultaneous gravimetric measurements of PM_{2.5} on PKU campus. A factor of 2.4 was achieved and used for DustTrak data correction, which was consistent with data reported in previous studies (Yanosky et al., 2002; Zhang and Zhu, 2010).

At Site B, a second set of water based CPC (Model 3785, TSI Inc., Shoreview, MN) and DustTrak (Model 8520, TSI Inc., Shoreview, MN) with a 2.5 μm inlet impactor were used to measure UFPs and PM $_{2.5}$, respectively. In addition, a Magee Scientific Aethalometer (Model AE-42, Magee Scientific Corporation, Berkeley, CA) was used to measure BC mass concentrations. The water based CPC was placed in a gate guard station and TSI conductive tubing was used to connect to the sampling inlet and extended through a window to outside ambient air. The DustTrak and Aethalometer were placed outdoor.

These two sets of instruments were operating simultaneously from 8 am to 9 pm during the sampling campaign. The CPCs were set to take readings every second. The DustTraks and the Aethalometer were set to measure at 1-min intervals. All instruments were within the manufacturer annual calibration. All the instruments were co-located before and after the field campaign to make sure there were no significant differences between the two sets of instruments in terms of real time readings. Data were downloaded each day immediately after the sampling was finished.

For traffic density, video recordings were made using a commercial camera with a tripod at the guard station. The camera was pointing towards the T intersection of North Zhongguancun Road and Chengfu Road. Recordings were made for 5 minutes every 15 minutes throughout the sampling period. Thus, for one hour of sampling, a total of 20 minutes recording were made. The number of vehicles was then counted per minute from the recording.

RESULTS AND DISCUSSION

UFP, PM_{2.5}, and BC Concentrations

The basic descriptive statistics for UFPs, PM_{2.5}, and BC concentrations are shown in Table 1. All of these measured concentrations show high variability. Comparing the data collected from Sites A and B, the average UFP number concentrations and the average PM_{2.5} mass concentrations

did not show significant differences. However, the standard deviation of UFP concentrations was much lower at Site A than at Site B (1.20×10^4 vs. 3.23×10^4 #/cm³). This indicates more stable UFP emission sources at Site A while the Site B UFP concentrations are primarily influenced by intermittent traffic emissions. The standard deviations of PM_{2.5} mass concentrations were similar at both sampling sites. PM_{2.5} was a little but not significantly higher (91.70 vs. $80.31 \, \mu g/m^3$) at Site A than at Site B. The measured average BC mass concentration at Site B was $4.70 \, \mu g/m^3$ with a standard deviation of $5.41 \, \mu g/m^3$ during the total 14 days of the sampling campaign. The high variability of BC mass concentrations at Site B is also likely due to the large number of diesel buses used for public transportation in Beijing.

At Site B, the average percentage of BC in PM_{2.5} was 5.9%. Literature values of BC and BC/PM_{2.5} ratios are listed in Table 3 and presented in Fig. 2. The reported average BC and PM_{2.5} concentrations at different sampling sites and at different sampling time are linearly correlated with an R² value of 0.87 (Fig. 2). The BC/PM_{2.5} ratio observed in this study was close to or slightly lower than most of those reported in previous studies conducted in Beijing (Dan et al., 2004; Yang et al., 2005; Zhao et al., 2013), as well as at other global sites. A one-year study at an urban site in Helsinky, Finland in 2000 through 2001 showed the BC in PM_{2.5} was about $14 \pm 8\%$ (Viidanoja et al., 2002). Another study conducted in New York state also showed a BC/PM_{2.5} mass concentration ratio of about 15% (Schwab et al., 2004). A previous study in Beijing in 2001 showed BC in PM_{2.5} was about 6.8% (He et al., 2001). This is likely because our data were mostly collected during daytime (from 8 am to 9 pm). A previous study conducted in Beijing urban and rural areas in 2009 indicated that heavy-duty diesel vehicle (HDDV) flow usually peaked around 240 vehicles/hour at midnight, but ranged from 0-20 vehicles/hour between 6 am and 9 pm. Accordingly, the diurnal BC concentrations reached the highest value of 29 µg/m³ at midnight in winter (Song et al., 2013). This is because due to the local traffic control rules in Beijing, HDDVs were only allowed to operate during night time (11 pm to 6 am next day) in the urban area. Thus, in this study, we were not able to capture the highest BC traffic emissions from those HDDVs. Since BC mass concentration is an indicator of combustion sources including diesel traffic, the lower percentage of BC in PM_{2.5} mass concentration in Beijing indicates that the diesel traffic may not be a predominant factor contributing to the total PM_{2.5} concentrations in winter in Beijing during the daytime.

Table 1. Basic descriptive statistics of UFP, PM_{2.5}, and BC.

	UFP (#/cm ³)		$PM_{2.5} (\mu g/m^3)$		BC (μ g/m ³)	
	Site A	Site B	Site A	Site B	Site B	
Mean	3.62×10^4	3.61×10^4	93.10	80.30	4.70	
Std Dev	1.20×10^{4}	3.23×10^{4}	91.71	87.02	5.41	
Geometric Mean	3.43×10^{4}	3.23×10^{4}	55.54	49.03	3.01	
Geometric Std Dev	1.38	1.59	2.87	2.66	2.62	
Min.	1.00×10^{3}	5.40×10^{3}	3.00	9.00	0.19	
Max.	1.13×10^{5}	2.20×10^{5}	5.85×10^{2}	5.43×10^{2}	29.43	

Correlations between UFP, PM_{2.5}, and BC

The UFP number concentrations and PM_{2.5} and BC mass concentrations were averaged at 30 minute intervals for all correlation analysis. Table 2 summarized Pearson's correlation coefficients among different pollutant measurements at both Site A and Site B. At both sampling sites, Pearson's correlations between UFPs and PM_{2.5} mass concentrations were not statistically significant. The UFP concentrations at Sites A and B were not significantly correlated either, despite that the average concentrations were within the similar range. This indicates the UFP sources at the two sampling sites were different albeit the two sites were only ~200 m apart. In contrast, Site B BC correlated with PM_{2.5} mass concentrations at Sites A and B, so did PM_{2.5} mass concentrations at both sampling sites (Table 2).

As shown in Fig. 2, data from previous studies have a strong spatial correlation between PM_{2.5} and BC mass concentrations. In the present study, a similarly high temporal correlation was observed between roadside BC and PM_{2.5} at both sampling sites. This result is not surprising since previous studies conducted in Beijing showed PM_{2.5} and BC share some common sources from coal and fuel burning, especially in the winter months (Dan *et al.*, 2004). Atmospheric scales of air pollutant transport can be roughly

categorized into microscale (0-100 meters), mesoscale (tens to hundreds of kilometers), synoptic scale (hundreds to thousands of kilometers) and global scale (> 10³ kilometers). Both PM_{2.5} and BC have been classified as mesoscale air pollutants that can travel tens to hundreds of kilometers (Krudysz et al., 2008; Wang et al., 2011). This is because their atmospheric lifetime is fairly long so that they can be transported and dispersed within a large region (Cape et al., 2012; Chen et al., 2013). When the atmospheric conditions are stagnant, PM_{2.5} and BC concentrations may also increase if local emission sources such as traffic exist. When the atmospheric PM_{2.5} levels are high, it generates the wellknown "Beijing Haze" phenomenon, which strongly affects visibility in the urban areas. Once PM_{2.5} and BC are emitted, their concentrations are affected by meteorological conditions such as wind speeds and directions. Furthermore, the vast majority of BC aerosol is in the fine particle size range (i.e., less than 2.5 μm) and typically comprises 5–10% of total $PM_{2.5}$ (Table 3).

Meteorological Effects on UFP and PM_{2.5}

In Beijing, local meteorology is often highly variable, which affects the local ambient air quality dramatically as reported in many previous studies (Feng *et al.*, 2005). Half-

Table 2. Pearson correlation coefficients among measured pollutants at campus site (A) and roadside site (B)

	UFP (A)	$PM_{2.5}(A)$	UFP (B)	PM _{2.5} (B)	BC (B)
UFP (A)	1.000(-)				
$PM_{2.5}(A)$	0.176 (0.002)	1.000(-)			
UFP (B)	0.318 (< 0.001)	0.166 (0.046)	1.000(-)		
$PM_{2.5}(B)$	0.012 (0.849)	0.824 (< 0.001)	0.037 (0.694)	1.000(-)	
BC (B)	0.150 (0.009)	0.871 (< 0.001)	0.008 (0.926)	0.753 (<0.001)	1.000(-)

Notes: P values are given in parentheses.

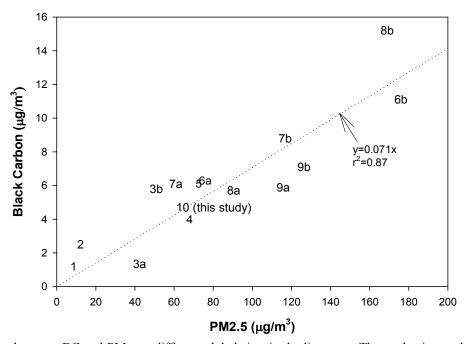


Fig. 2. Correlations between BC and PM_{2.5} at different global sites in the literature. The study sites and related references are listed in Table 3.

Study ID	Location	Year	BC Average (μg/m ³) (S.D.)	BC/PM _{2.5} (%)	Reference
1	Helsinki, Finland	2001-2002	1.2	14.0	Viidanoja et al. (2002)
2	Los Angeles, US	2011	2.5	20.6	Quiros <i>et al.</i> (2013)
3a	HongKong	2000-2001	Background: 1.36	3.2	Ho et al. (2002)
3b			Urban: 5.80	11.2	
4	Kaohsiung, Taiwan	1998-1999	4.0	5.9	Lin et al. (2001)
5	Pearl River Delta	2002	6.1	8.8	Cao et al. (2003)
6a	Beijing Urban	1999-2000	Summer: 6.3	8.3	He et al. (2001)
6b			Winter:11.1	6.3	
7a	Shanghai Urban	1999-2000	6.10	10.0	Yang et al. (2005)
7b	Beijing Urban	1999-2000	8.79	7.5	-
8a	Beijing Urban	2001-2003	Summer: 5.7 (2.9)	6.3	Dan et al. (2004)
8b			Winter: 15.2 (11.1)	9.0	
9a	Beijing Urban	2009	Summer: 5.9 (1.2)	5.1	Zhao et al. (2013)
9b		2010	Winter: 7.1 (2.2)	5.6	
10	Beijing Urban	2011	Winter: 4.7 (5.4)	5.9	This study

Table 3. Summary of literature values of BC and BC/PM_{2.5} ratio.

hour wind speeds and directions during the 14-day sampling campaign were summarized in Fig. 3 where the size of the dots indicates time of day and the distance from the center indicates different wind speeds. During the sampling periods, approximately 70% of the time, the wind blew from northwest to southeast (270–360 degrees), under which the Site A was upwind and Site B was downwind of the roads.

Collected data showed wind directions didn't significantly affect PM_{2.5} concentrations at both sites, BC concentrations at Site B, and UFP concentrations at Site B. Average UFP concentrations at Site A were slightly higher when the wind is from NW than other directions. Correlations between wind speeds and UFP and PM_{2.5} are shown in Fig. 4. To simplify the wind direction effects on the associations between wind speed and pollutants, only upwind UFP and PM_{2.5} data were used for Site A and downwind UFP and PM_{2.5} data were used for Site B. Wind speed and UFP number concentration did not show any correlations (Figs. 4(a) and 4(b)), however, wind speed and PM_{2.5} shows apparent log-decay correlations with R² values of 0.81 for the Site A and 0.80 for Site B (Figs. 4(c) and 4(d)). Higher wind speeds are associated with a fast drop in average PM_{2.5} mass concentrations at both sampling sites. This is consistent with previous findings that PM_{2.5} and BC share common regional sources such as biomass burning in winter in Beijing (Dan et al., 2004; Yang et al., 2005; Zhao et al., 2013). They also had comparable atmospheric lifetime on the order of weeks which make their concentrations affected similarly by air movements such as convection (atmospheric stability) and advection (wind), especially when regional source dominates.

Compared with PM_{2.5}, UFP concentrations tend to be indicative of local emissions. UFP concentrations were found to drop sharply as the distance from a roadway is increased due to both particle coagulation and atmospheric dispersion (Zhu *et al.*, 2002). However, the effect of wind speed on UFPs in this study is different from studies conducted in the US, where increasing wind speeds were shown to be statistically associated with a decrease in UFP

because of the dispersion effect from the air movement. In this study, the UFP concentrations remain relatively stable at different wind speeds. This is likely because in Beijing, extremely high PM_{2.5} levels can reduce UFP lifetime (Shi et al., 2007). In this study, much higher PM_{2.5} concentrations were observed at lower wind speeds (Figs. 4(c) and 4(d)). These fine particles provided larger surface areas for UFP to deposit. In addition, these fine particles provide a condensation sink for semi-volatile vapors and hence particle formation through nucleation processes both in vehicle exhaust plumes and in the wider atmosphere is suppressed by high pre-existing particle loadings. When the wind speed increased, assuming the emission rates were similar for all UFP sources, a stronger atmospheric dispersion was expected to reduce UFP concentrations. On the other hand, at higher wind speed, PM_{2.5} concentrations became much lower and the deposition surface for UFP became limited. These two competing processes may keep ambient UFP levels relatively stable with respect to wind speed in Beijing.

There is also a strong association ($R^2 = 0.96$) between wind speed and BC mass concentrations, with a profile similar to that between wind speed and $PM_{2.5}$. BC and $PM_{2.5}$ also share similar diurnal profile as shown in Fig. 5. This result is consistent with previous studies that have shown BC generally is enriched in the $PM_{2.5}$ size range (Chaloulakou *et al.*, 2003; Richmond-Bryant *et al.*, 2009).

Cooking Sources and Traffic Influence

Diurnal UFP, PM_{2.5}, and BC levels were analyzed against meteorological data. Wind speed was dichotomized into low wind (wind speed < 4.5 m/s) and strong wind (wind speed ≥ 4.5 m/s) as shown in Fig. 5. This results in about half and half data for each category. Under strong wind condition and when Site A was upwind of the roadways (wind is from 0 and 180 degrees), the UFP diurnal pattern shows bimodal at around 11 am and 6 pm (Fig. 5(b)). This result is consistent with previous study conducted on Tsinghua University campus, about 2 km from the sampling

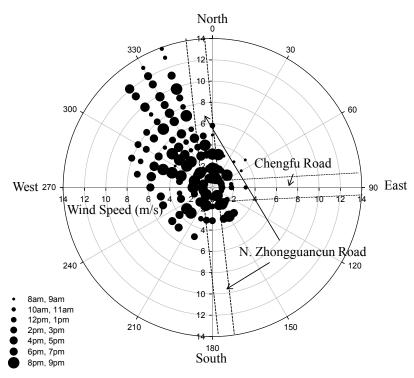


Fig. 3. Half-hour averaged wind speed and direction. Radius indicates wind speed and angle indicates wind direction, while 0 degree indicates north and 90 degree indicates east. Size of the dots on the graph indicates time of day with the smallest one representing 8–9 am.

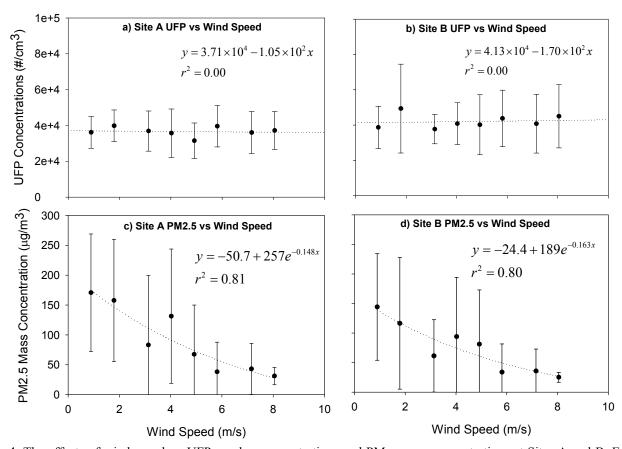


Fig. 4. The effects of wind speed on UFP number concentrations and $PM_{2.5}$ mass concentrations at Sites A and B. Error bars stand for one standard deviation.

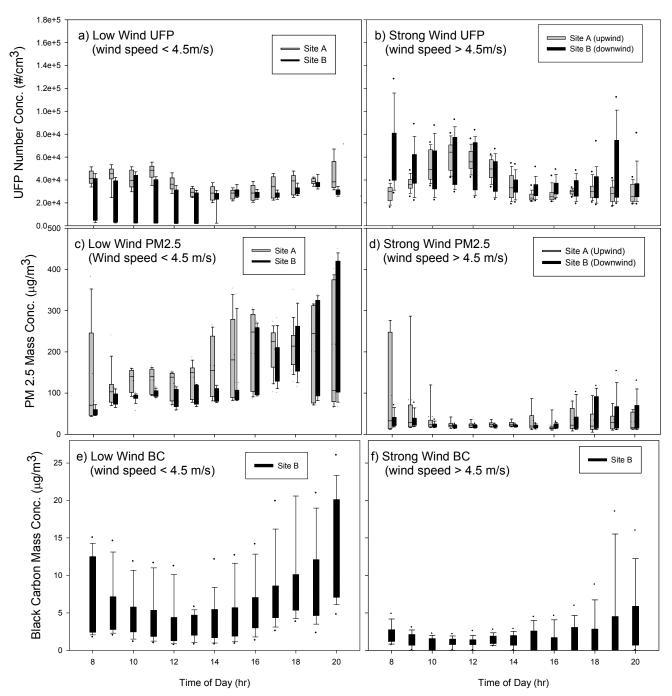


Fig. 5. Diurnal patterns of UFP, PM_{2.5}, and BC concentrations under low wind and strong wind conditions. The lines within boxes are median values. The boundaries of the boxes indicate 25^{th} and 75^{th} percentile of the data, whiskers indicate 10^{th} and 90^{th} percentile of the data, and the points indicate 5^{th} and 95^{th} percentile of the data.

sites used in this study (Shi *et al.*, 2007). Similar temporal profiles were reported in Beijing for hydrocarbon-like organic aerosol (HOA) which were related to cooking emissions (Zhang *et al.*, 2013). Previous studies have shown cooking emissions constitute approximately 20% of organic PM_{2.5} mass in Beijing (Huang *et al.*, 2006; Zhang *et al.*, 2014). At Site A, UFP temporal profile also suggests the influence from Chinese style cooking because the two modes were at approximately lunch and dinner hours, and there are many student dining halls located on campus as illustrated in Fig.

1(c). Most of the dining halls are located close to sampling Site A. One major dining hall (PKU Dining Hall) is about 100 meter southwest to Site A, and another one (Yannan Food) is about 200 meter west to Site A. There are many other dining halls on campus that are located within 500 meters from Site A. At Site B, UFP concentrations show bimodal at the morning and evening time during the traffic rush hours at early morning and late afternoon (Fig. 5(b)).

The average UFP number concentrations at Site A were similar during the low wind and strong wind time, but the

variations were greater on strong wind days (Figs. 5(a) and 5(b)). This indicates that stable atmosphere conditions allowed UFP to coagulate faster, but the strong winds made UFP disperse faster. In addition, under the low wind condition, PM_{2.5} levels are much higher which may provide more surface areas for UFP to deposit (Figs. 5(a) and 5(b)). In contrast to UFP number concentrations, average PM_{2.5} and BC mass concentrations were higher under low wind and had higher variations compared with strong wind. This diurnal pattern is consistent with the log-decay mode in Fig. 4 for the PM_{2.5} variations with wind speeds, especially during the daytime when the inversion effects are not strong and the atmosphere is relatively unstable (Figs. 5(c) and 5(d)). When the wind speeds were high, low variability of PM_{2.5} and BC was observed (Figs. 5(d) and 5(f)).

Traffic density is an important factor influencing roadside UFP levels. The HDVs (heavy duty vehicles), including buses and transport trucks but not heavy duty diesel trucks (HDDVs), comprise about 11.8% of the total traffic. The HDV vs. LDV (light duty vehicle) ratio was about 0.13. The traffic pattern on Chengfu Rd. shows that hourly traffic densities may become quite high during the morning and afternoon rush hours (8 AM–9 AM, 4 PM–7 PM, respectively), and is capable of reaching over 2500 vehicles per hour on average.

The overall hourly traffic densities were analyzed and compared with the corresponding hourly UFP concentrations at Site B. The analysis excluded data from periods when Site B was upwind of both Chengfu and N. Zhongguancun Roads. Average hourly UFP was found to have nearly no correlation with hourly total traffic densities ($R^2 = 0.05$). However, there was a relatively higher correlation between hourly UFP and HDV densities with an R^2 of 0.40.

CONCLUSIONS

In this study, traffic related pollutants including UFPs, PM_{2.5}, and BC were measured concurrently at a campus and a roadway sampling site near PKU campus from December 10^{th} through 23^{rd} , 2011. The traffic density data were collected with recorded video footage. Observed ambient UFP concentrations, PM2.5, and BC mass concentrations all showed high averages and strong variations at both sampling sites. The averages of UFP concentration and PM_{2.5} mass concentration at roadside were not significantly higher than campus site indicating local emission sources on campus. The regression results indicate strong wind speeds influenced PM_{2.5} and BC mass concentrations more than UFP concentrations. Temporal profiles of UFPs showed distinct impacts of traffic on roadway site measurements and Chinese style cooking as a potential source impacting campus site measurements. The high PM_{2.5} concentrations in Beijing might suppress UFP concentrations by providing a condensation sink for semivolatile vapors and extra surface areas for particle deposition.

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