

# PM<sub>1.0</sub> and PM<sub>2.5</sub> Characteristics in the Roadside Environment of Hong Kong

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Daily mass concentrations of PM<sub>1.0</sub> (particles less than 1.0 μm in diameter), PM<sub>2.5</sub> (particles less than 2.5 μm in diameter), organic carbon (OC), and elemental carbon (EC) were measured from January through May 2004 at a heavily trafficked sampling site in Hong Kong (PU). The average concentrations for PM<sub>1.0</sub> and PM<sub>2.5</sub> were 35.9 ± 12.4 μg cm<sup>-3</sup> and 52.3 ± 18.3 μg cm<sup>-3</sup>. Carbonaceous aerosols were the dominant species in fine particles, accounting for ~45.7% of PM<sub>1.0</sub> and ~44.4% of PM<sub>2.5</sub>. During the study period, seven fine-particle episodes occurred, due to the influence of long-range transport of air masses from mainland China. PM<sub>1.0</sub> and PM<sub>2.5</sub> responded in similar ways; i.e., with elevated mass and OC concentrations in those episode days. During the sampling period, PM<sub>1.0</sub> OC and EC generally behaved similarly to the carbonaceous aerosols in PM<sub>2.5</sub>, regardless of seasonal variations and influence by regional pollutions. The low and relatively constant OC/EC ratios in PM<sub>1.0</sub> and PM<sub>2.5</sub> indicated that vehicular emissions were major sources of carbonaceous aerosols. PM<sub>1.0</sub> and PM<sub>2.5</sub> had the same dominant sources of vehicular emissions in winter, while in spring PM<sub>2.5</sub> was more influenced by PM<sub>1–2.5</sub> (particles 1–2.5 μm in diameter) that did not form from vehicle exhausts. Therefore, PM<sub>1.0</sub> was a better indicator for vehicular emissions at the Roadside Station.

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## INTRODUCTION

In recent years, the problem of fine particles in urban areas has attracted increased concern, since the evidence from epidemiology and toxicology studies (Dockery et al. 1993; Schwartz et al. 1996; Katsouyanni et al. 2001) has suggested statistically significant association between morbidity and ambient fine-particle concentrations. Aerosol samples taken in urban areas show that motor vehicular emissions, especially diesel exhausts, usually constitute the most significant source of ultrafine (particle's aerodynamic diameter less than 0.1 μm) and fine particles (PM<sub>2.5</sub>) in urban environments (Zhu et al. 2002; Shi et al. 2001; Charron and Harrison 2003).

At present, Hong Kong Environmental Protection Department (HKEPD) recognizes that local, street-level pollution and the persistent regional smog problem are the most important air pollution issues for Hong Kong (<http://www.epd.gov.hk/>). Diesel vehicles are the main sources of street-level pollution. Smog, however, is caused by a combination of pollutants from motor vehicles, industry (e.g., power plants), vegetative burning and cooking, in both Hong Kong and the upwind area, the Pearl River Delta (PRD) regions.

Some consideration has been given in the United States to proposing that PM<sub>1.0</sub> be an ambient standard. The advantage of a PM<sub>1.0</sub> standard is not clear compared to the current PM<sub>2.5</sub> standard, but proposing it as a standard raises the question of whether PM<sub>1.0</sub> might provide better human health information than PM<sub>2.5</sub>. The answer is still controversial. Three different studies explored the relationship between PM<sub>1.0</sub> and PM<sub>2.5</sub> using MOUDI samplers in urban atmosphere in: Australia (Keywood et al. 1999); Helsinki, Finland (Vallius et al. 2000); and the Pittsburgh Supersite, USA (Cabada et al. 2004). All three studies concluded that the majority of PM<sub>1.0</sub> originated from the same sources as PM<sub>2.5</sub>, and measurement of PM<sub>1.0</sub> did not yield significant new information to add to that obtained from the PM<sub>2.5</sub> method. However, Lundgren et al. (1996), who conducted

the sampling with a trichotomous sampler in Phoenix, Arizona, USA, claimed that  $PM_{1.0}$  was a better indicator for a roadside microenvironment than  $PM_{2.5}$ , because, compared with  $PM_{2.5}$  and  $PM_{10}$ , it minimized interference from natural sources.

Past studies of  $PM_{10}$  (particle's aerodynamic diameter less than  $10\ \mu\text{m}$ ) (Yu et al. 2004; Qin et al. 1997) and  $PM_{2.5}$  (Cao et al. 2003; Ho et al. 2003; Louie et al. 2004; Louie et al. 2005) have provided limited detailed information on the roadside and/or regional pollution in Hong Kong. Few studies have investigated  $PM_{1.0}$  in Hong Kong, which is the dominant proportion of particulate matter in vehicular exhausts and transported air masses. In this study, daily 24-hour measurements of  $PM_{1.0}$  and  $PM_{2.5}$  were determined near a major road from January 21 to May 31, 2004 with two-collocated Partisol-Plus Model 2025 sequential air samplers (Rupprecht and Patashnick Co. Inc., Albany, NY). The sampling period was divided up into winter (January 21–February 29) and spring (March 1–May 31). Study objectives were to: (1) determine the levels of fine particles ( $PM_{1.0}$  and  $PM_{2.5}$ ) and carbonaceous aerosols in a roadside microenvironment; and (2) explore the relationship of  $PM_{1.0}$  and  $PM_{2.5}$  in a roadside microenvironment in various conditions, such as different seasons, regional pollution episodes and non-regional pollution days. The results are presented in this paper as mean value  $\pm$  standard deviation.

## METHODOLOGY

### Sampling Site

$PM_{1.0}$  and  $PM_{2.5}$  samples were collected at the PU Roadside Station air-quality monitoring site on the campus of the Hong Kong Polytechnic University (PU) (Figure 1), which is located

near Victoria Harbor. PU Roadside Station is in a residential and commercial area about 1–2 meters from the curb along Hong Chong Road, which leads to the Cross Harbor Tunnel. With four lanes in each direction, Hong Chong Road is one of the busiest cross-harbor roads in Hong Kong. Traffic flow is extremely high, with about 120,974 cars per day in both directions during sampling period (source: 2004 Annual Traffic Census in Hong Kong).

### Sample Collection

Daily  $PM_{1.0}$  and  $PM_{2.5}$  samples were acquired with two-collocated Partisol-Plus Model 2025 Sequential Air Sampler operated at  $16.7\ \text{l}\ \text{min}^{-1}$ . The particles were collected on 47 mm quartz fiber filters (QMA, Whatman International Ltd., Maidstone, England). They were weighed twice before and after sampling, using a Microbalance (Model MC5, Sartorius AG, Goettingen, Germany) with the sensitivity of  $\pm 1\ \mu\text{g}$  in the 0–250 mg range. Before weighing, the samples were equilibrated in a desiccator at  $20\text{--}30^\circ\text{C}$  and a relative humidity of 30–40% for 24 hours. Field blank filters were collected to reduce gravimetric bias due to filter handling during and/or after sampling. Louie et al. (2005) reported a 1:1 relationship based on a gravimetric analysis of collocated Teflon-membrane and quartz-fiber filters performed during a year-long sampling study in Hong Kong with the same instruments with this study. Similar results were also found for a study done in South Africa by Engelbrecht et al. (2001). However, large uncertainties were found to associate with the weighing of quartz-fiber filters by other studies. For example, Hitzenberger et al. (2004) reported  $\sim 38\%$  higher concentrations with quartz fiber filters than the average of other

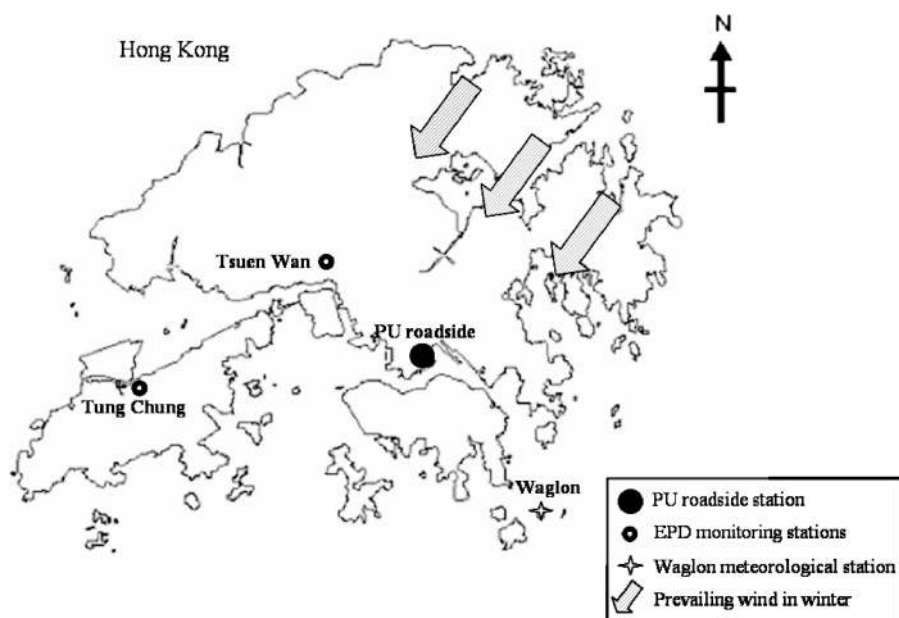


FIG. 1. Sampling locations for the Hong Kong Polytechnic University (PU) vehicle emissions dominated roadside station, two monitoring stations of Environmental Protection Department (EPD), and one meteorological station.

methods. Therefore the mass concentrations in the present study maybe have uncertainty although the filters were handled very carefully during the study.

Before sampling, quartz fiber filters were preheated in an electric furnace at 900°C for at least three hours in order to remove carbonaceous contaminants. The filters were handled only with tweezers cleaned with dry KimWipes (Kimberly-Clark Corporation, USA) to reduce the possibility of contamination. After weighing, samples were stored in a refrigerator at about 4°C before chemical analysis to prevent the evaporation of volatile components.

### Analysis of Organic and Elemental Carbon

The samples were analyzed for OC and EC using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (AtmAA Inc, Calabasas, CA, USA) with the IMPROVE thermal/optical reflectance (TOR) protocol (Chow et al. 1993). The DRI Model 2001 carbon analyzer (Chow et al. 2004) is based on the preferential oxidation of OC and EC compounds at different temperatures. It relies on the fact that OC is volatilized from the sample deposit in a helium (He) atmosphere at low temperatures, while EC is not consumed. The TOR protocol heats the sample (0.526 cm<sup>2</sup> per punch) stepwise at temperatures of 120°C (OC<sub>1</sub>), 250°C (OC<sub>2</sub>), 450°C (OC<sub>3</sub>), and 550°C (OC<sub>4</sub>) in a non-oxidizing He atmosphere, and 550°C (EC<sub>1</sub>), 700°C (EC<sub>2</sub>), and 800°C (EC<sub>3</sub>) in an oxidizing atmosphere with 2% oxygen (O<sub>2</sub>) in He. The evolved carbon is oxidized into carbon oxide (CO<sub>2</sub>) and then reduced to methane (CH<sub>4</sub>) for quantification with a flame ionization detector (FID). The pyrolysis of OC is continuously monitored by a Helium-neon (He-Ne) laser at a wavelength of 632.8 nm. OC is defined as the portion of carbon evolved before the temperature at which the filter reflectance resumes the initial level. The carbon evolved beyond this temperature is defined as EC. The minimum detection limit (MDL) of carbon combustion methods is 0.82 μg C cm<sup>-2</sup> for OC, 0.19 μg C cm<sup>-2</sup> for EC, and 0.93 μg C cm<sup>-2</sup> for total carbon (TC, sum of OC and EC). All samples in this study have concentrations higher than MDL. Replicated analyses were performed for ~10% of all samples. Also, field blank filters were collected to provide information about contaminants that may be introduced during sample collection, storage, and transport.

The sampling with quartz filters is prone to positive and negative artifacts that result in erroneous measurements of particulate organic carbon (Turpin et al. 1994; Kim et al. 2001; Watson and Chow 2002; Subramanian et al. 2004). The OC artifacts are not determined in the present study since the instrument is not suitable for this measurement. Positive 24-hour OC artifacts were estimated at the same sampling location using one URG sampler (URG-3000ABC, Chapel Hill, USA) in winter (December 2004–January 2005) and summer (July 2005–September 2005). The URG sampler is able to collect PM<sub>1.0</sub> and PM<sub>2.5</sub> simultaneously. The positive OC artifacts were estimated with a backup quartz filter placed behind the main quartz filter because the

quartz behind quartz (QBQ) approach provides a reasonable estimate of the positive artifacts on the bare quartz filter for the 24-hour samples (Subramanian et al. 2004). The positive OC artifacts (QBQ) in winter were found to be almost consistent for PM<sub>1.0</sub> and PM<sub>2.5</sub>,  $1.8 \pm 0.5 \mu\text{g C m}^{-3}$  and  $1.9 \pm 0.5 \mu\text{g C m}^{-3}$ , respectively. In summer, the OC artifacts were  $1.5 \pm 0.3 \mu\text{g C m}^{-3}$  for PM<sub>1.0</sub> and  $1.6 \pm 0.4 \mu\text{g C m}^{-3}$  for PM<sub>2.5</sub>. Negative artifacts due to volatilization of particle-phase organics from the particle sample were not quantified, because of limited resources.

### EPD PM<sub>2.5</sub> Monitoring Station

Tsuen Wan and Tung Chung are typical urban, ambient monitoring stations (Figure 1) operated by HKEPD. PM<sub>2.5</sub> data measured at these two stations were used to evaluate the relationship of fine particles among different locations, such as ambient and roadside site. Equipment at these sampling sites had adequate exposure, with monitoring probes placed 17 and 21 m above ground level, located on rooftops of buildings. Road dust from nearby streets does not contribute appreciably at this elevation (Louie et al. 2002).

Hourly PM<sub>2.5</sub> mass at Tsuen Wan and Tung Chung stations was measured with the Series 1400a PM<sub>2.5</sub> Tapered Element Oscillating Microbalance monitor (TEOM, Rupprecht & Patashnick Co., Inc., Albany, NY, USA), which is a true “gravimetric” instrument that continuously weighs the Teflon-coated borosilicate glass filter, and calculates near real-time PM<sub>2.5</sub> mass concentrations. The sample stream is preheated to 50°C before entering the mass transducer to eliminate the necessity for humidity equilibration for the hydrophobic filter.

### Meteorological Parameters during Sampling Periods

Hong Kong's climate is sub-tropical with four seasons and influenced by the Asian monsoon. In summer, the prevailing wind is southerly, bringing to Hong Kong clean marine air masses. In winter, continental emissions from interior Asia intrude into Hong Kong and the South China Sea, accompanying a prevailing northeasterly wind. Therefore, Hong Kong's air quality during winter is influenced by local and regional sources. Spring and autumn are transit seasons.

Throughout the sampling periods, the mean air pressure, temperature, and relative humidity did not vary significantly. During wintertime, the prevailing surface wind direction was northeasterly with ~82% of hourly winds from the resultant vector of 38°, as shown in Figure 2a. This means that nearly all winter winds traveled over China's continental land mass before reaching Hong Kong. During springtime, however, the mean directions of hourly surface winds varied in all vectors, as displayed in Figure 2b. The resultant vector is 77°, accounting for ~48% of wind distribution. Wind from the south (90°–270°) was ~38% of distribution. Wind-rose analysis suggests that the air masses arriving at Hong Kong in spring were not only from the continent, but sometimes also from the South China Sea. The above

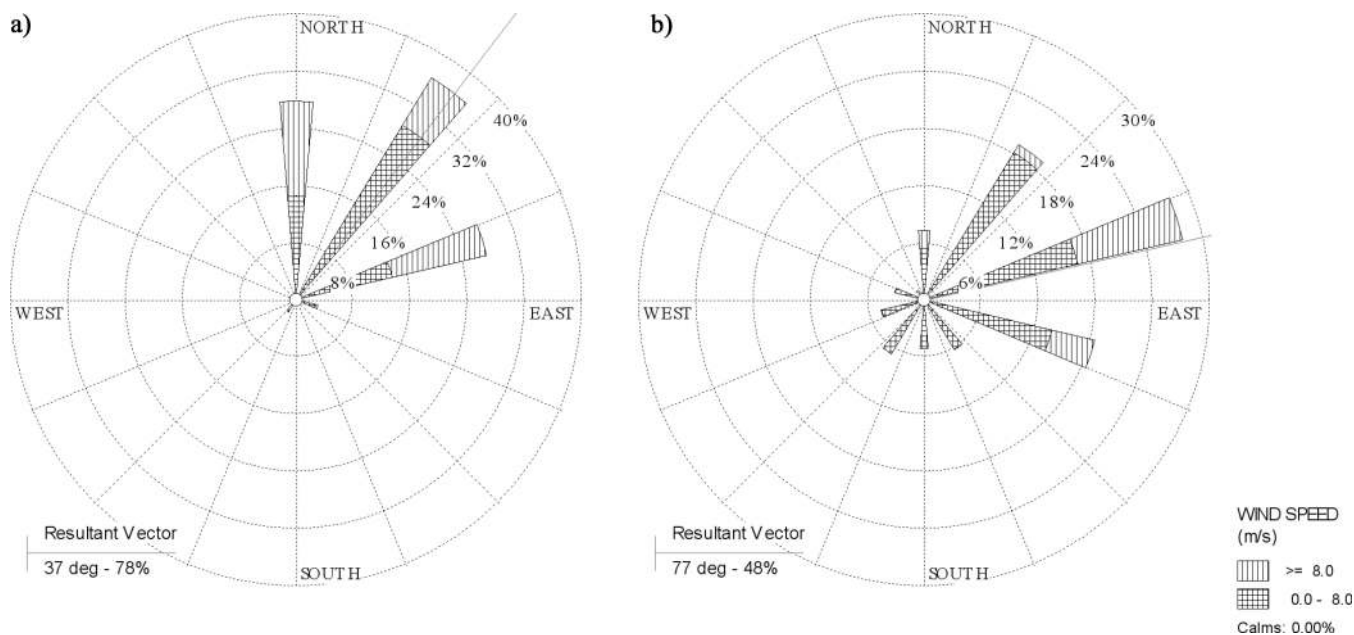


FIG. 2. Frequency distribution of surface wind in winter (a) and in spring (b).

weather parameters were measured at Waglon Meteorological Station (22.18°N, 114.30°E) of Hong Kong Observation, which is located on Waglon Island in an open landscape (Figure 1).

## DISCUSSION

### Concentrations of Mass, Organic Carbon, and Elemental Carbon

Table 1 summarizes the  $PM_{2.5}$  mass and carbon concentrations. On average, the mass concentrations of  $PM_{1.0}$  and particulate at PU Roadside Station for all 132 sampling days were  $35.9 \pm 12.4 \mu\text{g m}^{-3}$  and  $52.3 \pm 18.3 \mu\text{g m}^{-3}$ , respectively. The range of particle concentrations varied significantly, from  $17.5 \mu\text{g m}^{-3}$  to  $85.0 \mu\text{g m}^{-3}$  for  $PM_{1.0}$  and from  $24.3 \mu\text{g m}^{-3}$  to  $111.4 \mu\text{g m}^{-3}$  for  $PM_{2.5}$ , respectively. Overall  $PM_{1.0}$  consisted of  $\sim 19.4\%$  of OC and  $\sim 26.4\%$  of EC, and for  $PM_{2.5}$ ,  $\sim 21.1\%$  of OC and  $\sim 23.4\%$  of EC. The average OC/EC ratio was 0.8 and 1.0 for  $PM_{1.0}$  and  $PM_{2.5}$ , respectively.

Table 2 lists OC and EC levels from several past studies. Since there is no international uniform analysis method, different thermal methods were used in some studies, which might result in large variations in OC and EC measurements. Li and Lin (2002) measured the carbon concentrations in  $PM_{1.0}$  via a combustion technique in Taipei, Taiwan during October of 1999. The average OC/EC ratio of 1 at that traffic site in Taipei was similar to averages in this study, but generally, average OC/EC ratio in urban ambient atmospheres in Taipei is higher than roadside ratios in Taipei and in this study. For  $PM_{2.5}$ , EC measured at the PU Roadside site exceeded the average  $PM_{2.5}$  EC concentration found in a previous study (Cao et al. 2003) by a factor of  $\sim 1.0$ . This is due to the different distances the sampler was placed from the road in each study, about 1–2 meters for this study and more than 30 meters for the study of Cao et al. (2003). Zhu et al. (2002) observed a dramatic decrease in concentrations of ultrafine particles and BC near the 710 freeway in Los Angeles, California, USA when the sampler was moved from 4 meters to 17 meters away from curbside. This suggests that the increased EC levels

TABLE 1  
Seasonal average of  $PM_{1.0}$  and  $PM_{2.5}$  mass and carbonaceous aerosols at PU Roadside Station

	PU roadside					
	$PM_{1.0}$			$PM_{2.5}$		
	Mass ( $\mu\text{g m}^{-3}$ )	OC ( $\mu\text{g C m}^{-3}$ )	EC ( $\mu\text{g C m}^{-3}$ )	Mass ( $\mu\text{g m}^{-3}$ )	OC ( $\mu\text{g C m}^{-3}$ )	EC ( $\mu\text{g C m}^{-3}$ )
Winter	$40.9 \pm 15.5$	$8.3 \pm 3.3$	$9.0 \pm 3.8$	$54.1 \pm 21.1$	$13.2 \pm 6.0$	$11.0 \pm 4.7$
Spring	$33.8 \pm 10.1$	$6.4 \pm 1.9$	$9.7 \pm 3.3$	$51.5 \pm 17.0$	$10.0 \pm 3.7$	$12.7 \pm 4.2$
Total	$35.9 \pm 12.4$	$7.0 \pm 2.6$	$9.5 \pm 3.5$	$52.3 \pm 18.3$	$11.0 \pm 4.7$	$12.2 \pm 4.4$

TABLE 2  
Levels of carbonaceous aerosols in urban atmosphere in Asia

Location	Sampling period	Size	$(\mu\text{g C m}^{-3})$				Location	Method	Reference
			TC	OC	EC	OC/EC			
Hong Kong, PU	2004.1-2	PM <sub>1.0</sub>	17.3	8.3 ± 3.3	9.0 ± 3.8	1.0	Roadside <sup>a</sup>	IMPROVE-TOR	This study
Tai Wan, Taipei	1999.10	PM <sub>1.0</sub>	22.9	11.5 ± 2.8	11.3 ± 1.7	1.0	Roadside	Combustion	Li C.S. 2002
Tai Wan, Taipei	1999.12	PM <sub>1.0</sub>	4.7	3.4 ± 0.9	1.3 ± 0.7	2.6	Urban	Combustion	Li C.S. 2002
Hong Kong, PU	2004.1-2	PM <sub>2.5</sub>	24.2	13.2 ± 6.0	11.0 ± 4.7	1.3	Roadside <sup>a</sup>	IMPROVE-TOR	This study
Hong Kong, PU	2002.1-2	PM <sub>2.5</sub>	16.7	10.6 ± 3.7	6.1 ± 1.8	1.7	Roadside <sup>b</sup>	IMPROVE-TOR	Cao J.J. 2003
Hong Kong, MK	2000.11-2001.10	PM <sub>2.5</sub>	36.8	16.7 ± 7.6	20.2 ± 4.2	0.8	Roadside <sup>a</sup>	IMPROVE-TOR	Louie P. 2004
Hong Kong, TW	2000.11-2001.10	PM <sub>2.5</sub>	14.1	8.7 ± 5.3	5.4 ± 1.4	1.6	Urban	IMPROVE-TOR	Louie P. 2004
Hong Kong, HT	2000.11-2001.10	PM <sub>2.5</sub>	5.9	4.2 ± 3.7	1.7 ± 0.9	2.5	Rural	IMPROVE-TOR	Louie P. 2004
China, GZ	2002.1-2	PM <sub>2.5</sub>	23.8	17.8 ± 10.2	6.0 ± 3.2	2.9	Urban	IMPROVE-TOR	Cao J.J. 2003
China, SZ	2002.1-2	PM <sub>2.5</sub>	19.2	13.2 ± 4.1	6.1 ± 1.8	2.2	Urban	IMPROVE-TOR	Cao J.J. 2003
China, ZH	2002.1-2	PM <sub>2.5</sub>	17.3	12.2 ± 4.4	5.0 ± 1.6	2.4	Urban	IMPROVE-TOR	Cao J.J. 2003
Korea, Chongju	1995, Winter	PM <sub>2.5</sub>	9.3	5.0	4.3	1.2	Urban	IMPROVE-TOR	Lee H. S. 2001

<sup>a</sup>1–2 m away from the major road, <sup>b</sup>more than 30 m away from the major road.

in this study were mainly attributed to the sampler being much closer to road. Table 2 also shows the PM<sub>2.5</sub> OC/EC results for other sites where the TOR method was used to conduct OC/EC analyses. Generally, the carbonaceous aerosols (TC) in PRD region, including Guangzhou (GZ), Shenzhen (SZ), Zhuhai (ZH) (Cao et al. 2003) and Hong Kong (Louie et al. 2004), were higher than Chongju, Korea (Lee et al. 2001). High OC concentrations, observed in the urban atmosphere of GZ, SZ, and ZH in China, formed potential contamination sources for Hong Kong during the prevailing Asian monsoon (winter). In Hong Kong, the rural OC/EC ratio was similar to other cities in the PRD region (Cao et al. 2003), followed by urban and roadside ratios.

OC concentrations in PM<sub>1.0</sub> and PM<sub>2.5</sub> at PU Roadside Station showed a clear, seasonal pattern ( $p$ -value < 0.05 for both of PM<sub>1.0</sub> and PM<sub>2.5</sub>, two-tailed  $t$ -test) with ~30% higher concentrations in winter than spring (Table 1). Regional or long-range transport of continental aerosols was believed to be the dominating factor leading to higher OC levels in Hong Kong during winter because vehicles emissions would not have significant day-to-day variations due to the consistent traffic flow in each day (2004 Annual Traffic Census). Several pollutants in the Hong Kong atmosphere have been affected by long-range transport of continental aerosols, which lead to higher pollution levels in winter; more than other seasons (Pathak et al. 2003; Louie et al. 2005; Yu et al. 2004). Most of these pollutants are secondary aerosols, such as OC (Louie et al. 2005; Yu et al. 2004), and sulphate and ammonium (Pathak et al. 2003), that were thought to be produced by gas-to-particle conversion, or chemical reaction, during transport. In a study done by Pathak et al. (2003), continental long-range transport of aerosols was found to increase sulfate and ammonium concentrations in Hong Kong's air by 49% to 383%, and 33% to 302%, in 2000–2001.

EC originates from relatively simple sources and does not form in atmosphere due to its nearly inert property (Ogren and Charlson 1983). It mainly originates from incomplete combustion of carbon-containing material (Ogren and Charlson 1983). In this study, EC seems controlled by local sources, such as vehicular exhausts, that continuously emit EC into the atmosphere. This results in weak seasonal variations, as seen in Table 1. Besides vehicular emissions, Yu et al. (2004) found that ship emissions at Hong Kong's container port were also important contributors to EC loadings. In this study, the PU roadside station was upwind of the nearby port (Victoria Harbor) in winter, so the effects from ship emissions were limited. In spring, however, according to Yu et al. (2004), the PU Roadside might have received the plume from ship emissions when the wind direction was from the south. Yu et al. (2004) claimed that EC seasonality in Hong Kong was more dependent upon the distance from, and relative location to, the city's container port.

### Regional Pollution Episodes

As showed in Figure 3, time series of PM<sub>1.0</sub> exhibited seven episode days, January 30, February 14–15, February 23, February 26, April 19–20, respectively, during which PM<sub>1.0</sub> exceeded the average concentration by a factor of two. It was found that the PM occurring during episode days differed statistically from non-episode days ( $p$ -values < 0.05,  $t$ -test). Concurrent peaks for PM<sub>2.5</sub> were also found at PU Roadside Station and urban ambient monitoring stations (Tung Chung and Tsuen Wan). For instance, the average PM<sub>2.5</sub> loading during the episodes at Tsuen Wan exceeded the average level throughout the study period by a factor of 2.4.

Five-day back trajectories were conducted for those episode days using a HYSPLIT model (Draxler and Hess 1997). All of

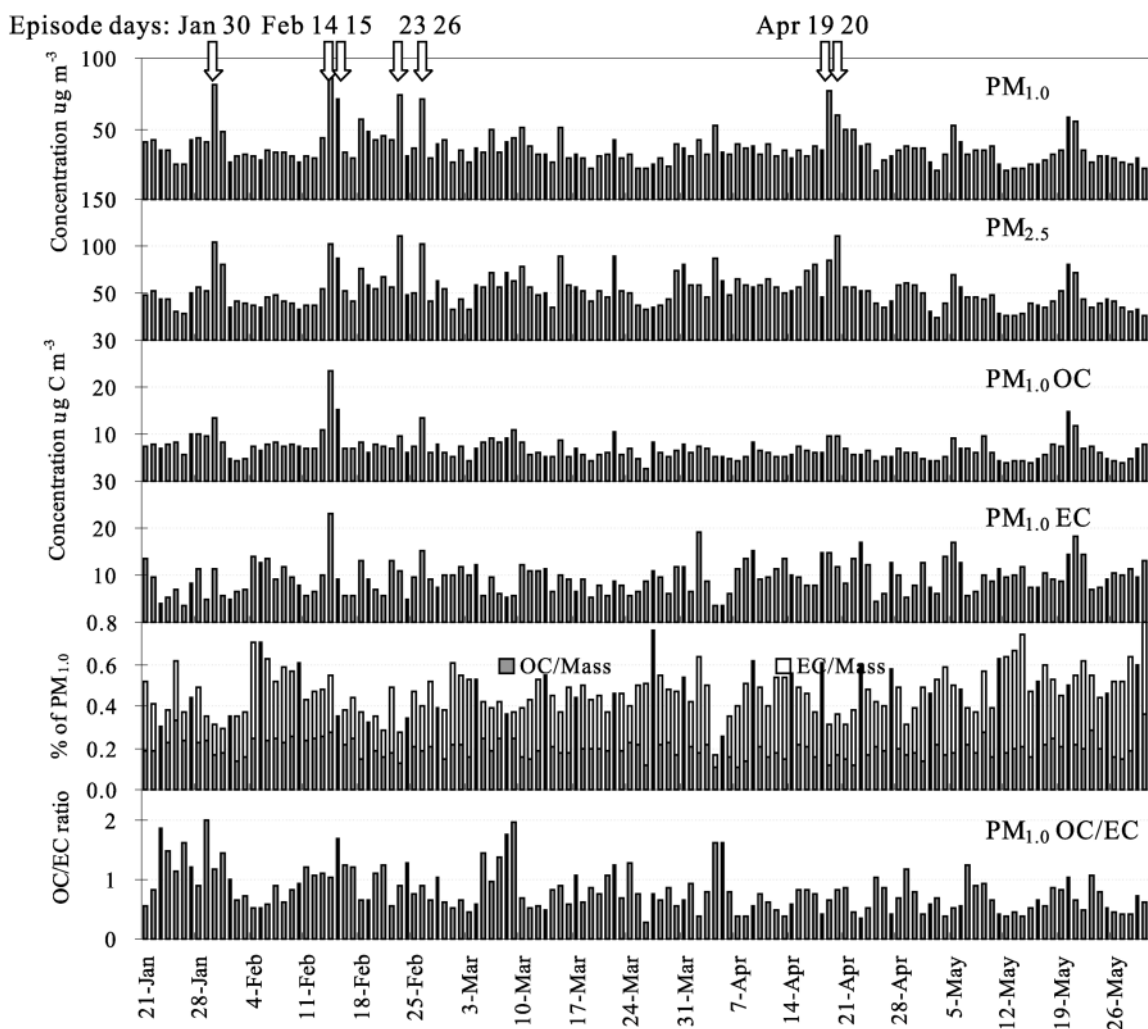


FIG. 3. Daily 24-h  $PM_{1.0}$ ,  $PM_{2.5}$ ,  $PM_{1.0}$  OC,  $PM_{1.0}$  EC concentrations, and OC/mass, EC/mass, OC/EC ratios in  $PM_{1.0}$  at PU roadside during January to May 2004.

the air masses during episode days originated from north China, traveling across southeastern China along the coastline before reaching Hong Kong and bringing aged, polluted aerosols. Similar pollution episodes have been found in prior studies (Yu et al. 2004; Louie et al. 2005).

Regression analysis in Figure 4a shows that  $PM_{2.5}$  collected at Tsuen Wan and Tung Chung generally had good relationship with a correlation coefficient ( $R$ ) equal to 0.86 in non-episode days, and 0.90 during episode days. This indicates that they are controlled by similar urban- and regional-scale sources, including remote sources. The particulate spatial distribution tended to be more even in the ambient atmosphere, due to the effects of remote sources. As seen in Figure 4b, PU Roadside  $PM_{2.5}$  moderately correlated with  $PM_{2.5}$  at Tsuen Wan in non-episode days ( $R = 0.75$ ), implying that vehicular emissions, as an urban plume, had contributed to urban ambient  $PM_{2.5}$  during non-episode days. Poor relationship ( $R = 0.28$ ) was found during episode days, which suggests that particles at PU Roadside were

comprised from sources that do not contribute much to urban atmosphere. Most likely, nearby vehicular emissions were those sources.

Occurrence of episodes in Hong Kong during winter was found to associate with a mesoscale subsiding airstream with a moderate to stagnant easterly transport (Louie et al. 2005). In this study, the average mixing height during episode days was 666 m, much lower than average mixing height during the non-episode periods (936 m). This indicated poor dispersion conditions when the episodes occurred. Thus, the elevated concentrations of pollutants observed at PU during episode days were attributed to the combination of stagnant vehicular emissions from the nearby road, and subsiding continental aerosols from long-range transport.

Table 3 shows concentrations of carbonaceous aerosols for each episode day. Carbonaceous aerosols on 14 February were found to be much higher than on other episode days (Table 3), comprised of  $\sim 67.4\%$  of  $PM_{1.0}$  and  $\sim 54.3\%$  of  $PM_{2.5}$ . The

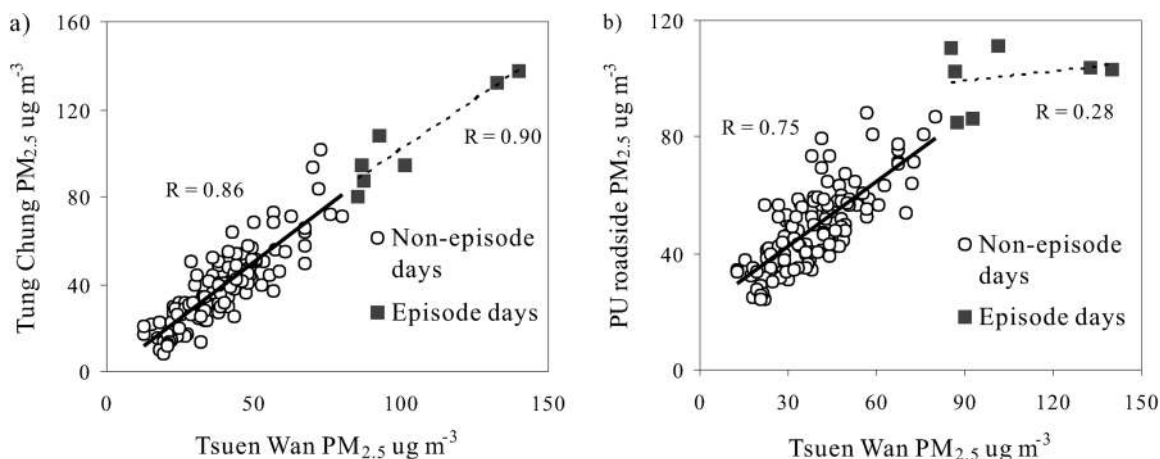


FIG. 4. Correlations of  $PM_{2.5}$  mass between Tsuen Wan and Tung Chung (a) as well as Tsuen Wan and PU Roadside Station (b) during January to May 2004.

sample may have been contaminated from a heavy-duty truck that occasionally idled near the sampling location during sampling period. Therefore, the calculation of average values for episode days excludes the data from 14 February.

On episode days, the average OC concentration in  $PM_{1.0}$  and  $PM_{2.5}$  increased  $\sim 70\%$  and  $\sim 100\%$ , respectively, compared to average values. EC showed only a 20–30% increase. The fact that higher concentrations of OC occurred during episode days confirmed our previous assumption that the seasonal pattern of high concentration in winter and low concentration in spring was attributed to the long-range transport of continental aerosols. During episode days, total carbonaceous aerosols ( $TC = OC + EC$ ) accounted for  $\sim 33.4\%$  and  $\sim 37.3\%$  of  $PM_{1.0}$  and  $PM_{2.5}$ , respectively (Table 3), which were lower than aver-

age contributions in sampling period,  $\sim 45.7\%$  and  $\sim 44.4\%$  of TC in  $PM_{1.0}$  and  $PM_{2.5}$ , respectively. All of this shows that particles at PU Roadside were influenced by remote sources that had chemical compositions other than those from primary vehicle emissions. Further study should be conducted on the chemical characterization on episode days in the future in order to evaluate the influence of long-range transport of continental aerosols on Hong Kong.

#### Relationships between $PM_{1.0}$ and $PM_{2.5}$

Particles with different size fractions possibly differ not only in size and morphology, but also in: formation mechanisms; sources; and, chemical and physical properties. In this study, regression analysis was used to evaluate the relationships between

TABLE 3  
Summary statistics for the concentrations of PM, carbonaceous aerosols, and ratios during episode days in 2004

	Episode days						
	30 Jan	14 Feb <sup>a</sup>	15 Feb	23 Feb	26 Feb	19 Apr	20 Apr
<b><math>PM_{1.0}</math></b>							
Mass $\mu g m^{-3}$	80.6	85.0	71.0	74.6	70.9	77.3	59.0
OC $\mu g C m^{-3}$	13.5	23.3	15.4	9.5	13.4	9.4	9.6
EC $\mu g C m^{-3}$	11.5	22.9	9.2	10.8	15.1	14.8	11.6
OC/EC	1.2	1.0	1.7	0.9	0.9	0.6	0.8
OC/ $PM_{1.0}$ %	16.7	27.4	21.7	12.7	18.9	12.2	16.3
EC/ $PM_{1.0}$ %	14.3	26.9	13.0	14.5	21.3	19.1	19.7
<b><math>PM_{2.5}</math></b>							
Mass $\mu g m^{-3}$	103.7	103.1	86.5	111.4	102.3	84.8	110.8
OC $\mu g C m^{-3}$	23.2	35.8	21.1	21.9	30.6	13.9	22.4
EC $\mu g C m^{-3}$	13.9	28.6	11.5	10.9	18.7	15.0	20.3
OC/EC	1.7	1.3	1.8	2.0	1.6	0.9	1.1
OC/ $PM_{2.5}$ %	22.4	34.7	24.4	19.7	30.0	16.3	20.2
EC/ $PM_{2.5}$ %	13.4	27.8	13.3	9.7	18.3	17.6	18.3

<sup>a</sup>The sample might be contaminated.

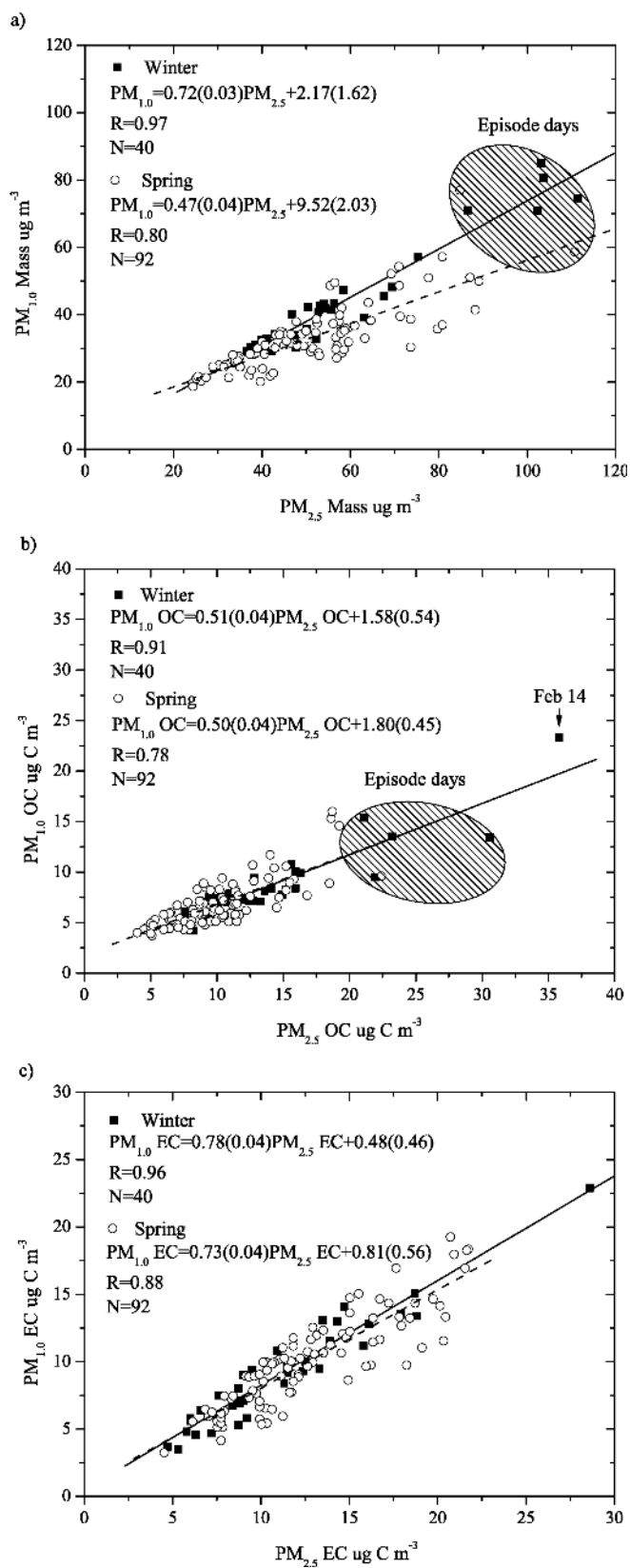


FIG. 5. Correlations between  $PM_{1.0}$  and  $PM_{2.5}$  for mass (a), OC (b), and EC (c) (Standard error is in the bracket).

$PM_{1.0}$  and  $PM_{2.5}$  (Figure 5), which is the most common method used in earlier studies (Keyword et al. 1999; Vallius et al. 2000; Cabada et al. 2004). The data derived on episode days was highlighted to differentiate from non-episode days (Figures 5a and 5b).

As shown in Figures 5b and 5c, the behavior of carbonaceous aerosols (OC and EC) in  $PM_{1.0}$  generally followed those in  $PM_{2.5}$  whether it was winter or spring. Meanwhile, the OC/EC ratios were low and relatively constant for both  $PM_{1.0}$  and  $PM_{2.5}$ , averaging  $0.8 \pm 0.4$  for  $PM_{1.0}$  and  $1.0 \pm 0.4$  for  $PM_{2.5}$ . This suggests that nearby vehicular emissions were the major sources for carbonaceous aerosols.

As illustrated in Figure 5a,  $PM_{1.0}$  and  $PM_{2.5}$  were highly correlated in winter regardless of episodes, with a correlation coefficient of 0.97 indicating they were controlled by similar sources; most likely vehicular emissions, in this case. In contrast, the trend of regression fitting curve in spring did not follow that in winter. The contribution of  $PM_{1-2.5}$  to  $PM_{2.5}$  seemed to have increased, especially for high concentrations. The correlation coefficient between  $PM_{1-2.5}$  and  $PM_{2.5}$  also increased from 0.85 in winter to 0.91 in spring. Furthermore, the difference resulted mainly from non-carbonaceous aerosols, indicating the contribution of non-vehicular emissions. Thus  $PM_{1.0}$  seems to be a better indicator for vehicular emissions at the PU Roadside microenvironment than  $PM_{2.5}$ , because, compared  $PM_{2.5}$ , it was not as greatly influenced by non-vehicle sources.

## CONCLUSION

Aerosol fine-particle samples ( $PM_{1.0}$  and  $PM_{2.5}$ ) were collected daily during winter and spring along the heavily trafficked Chong Hong Road in Hong Kong (PU Roadside Station), for nearly five months. An integrated data set, including aerosol mass and concentrations of carbonaceous aerosols associated with episode and non-episode days, has been obtained. The information obtained is especially important for helping to enhance the current understanding of two important air pollution problems confronting Hong Kong—local street pollution and regional smog pollution.

$PM_{1.0}$  and  $PM_{2.5}$  measurements at PU Roadside showed that carbonaceous aerosols were major components in fine particles, constituting  $\sim 45.7\%$  of  $PM_{1.0}$  and  $\sim 44.4\%$  of  $PM_{2.5}$ . Particle mass and OC showed higher concentrations in winter than spring, due to the contribution of continental aerosol pollutants brought in by long-range transport.

Seven fine particle episodes occurred during the study period. At PU Roadside Station,  $PM_{1.0}$  and  $PM_{2.5}$  responded in similar ways during these episode days, with elevated mass and OC concentrations. The contribution of carbonaceous aerosols to particulate mass was lower during episode days than non-episode days.

Regression analysis shows that  $PM_{1.0}$  OC and EC generally behaved similarly to  $PM_{2.5}$ . Vehicular emissions were a major source of both, although OC concentration increased during win-



ter due to influences from remote sources. During winter, PM<sub>1.0</sub> and PM<sub>2.5</sub> originated from the same major sources (vehicular emissions) regardless of episodes, while PM<sub>2.5</sub> was more influenced by PM<sub>1–2.5</sub> that obviously did not form from vehicular emissions. PM<sub>1.0</sub>, therefore, was a better indicator for vehicular emissions at PU Roadside Station.

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