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ATMOSPHERIC AEROSOLS DURING POLLUTION EPISODES IN HONG KONG

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ABSTRACT

The highest frequency of pollution episodes occurred in winter and spring in Hong Kong. Time series for $PM_{1.0}$, $PM_{2.5}$ and the concentrations of carbonaceous aerosols were determined for samples collected from a heavily-traffic road in Hong Kong from January 21 to May 31, 2004. Seven episode days for $PM_{1.0}$ and $PM_{2.5}$ were observed during the whole measuring period with much higher fine particle loadings ($PM_{1.0}$ and $PM_{2.5}$) exceeding the observed average concentration by a factor of two. Of the six episode days (one susceptive sample was exclusive), four days were in winter with heavy northeasterly wind, the other two days were in spring with a low wind speed from south. During the six episode days, elevated OC and EC concentrations were also observed, while the total carbonaceous aerosols only accounted for ~33% for $PM_{1.0}$ and ~37% for $PM_{2.5}$, respectively. In contrast, it was founded that carbonaceous aerosols accounted for ~88% and ~78% for $PM_{1.0}$ and $PM_{2.5}$ when local street-level pollution was dominated source in spring. The correspondence of $PM_{1.0}$ to regional episodes seemed much sensitive than that of $PM_{2.5}$. The effect of wet removal mechanism was found to be more effective for fine particle and OC in winter possibly due to the presence of soluble secondary aerosols (e. g. polar organic materials and sulfates) in aged air mass transported to Hong Kong.

Key Words: Organic Carbon (OC); Elemental Carbon (EC); $PM_{1.0}$; $PM_{2.5}$

1. INTRODUCTION

In recent years, the problem of fine particles in urban area has attracted increased concerns since the evidence from epidemiology and toxicology (Dockery et al. 1993; Schwartz et al. 1996; Katsouyanni et al. 2001) has suggested statistical significant association between morbidity and ambient fine particle concentrations. Aerosol samples taken in urban areas show that motor vehicular emissions, especially for diesel exhausts, usually constitute the most significant source of ultrafine and fine

particles in an urban environment (Zhu et al. 2002; Shi et al. 2001; Charron and Harrison, 2003). Therefore it is important to understand the levels of fine particles and regular variation near roadways especially regarding the public health of those traveling on and living in close proximity to roads.

Hong Kong (22.12N, 114.08E) is located on the southeastern coast of the Asiatic mainland with quite mountainous and some terrain changes rapidly from sea level to an elevation of several hundred meters or higher. The population spreads unevenly over this region. The sampling site in this study located in the most residential and commercial areas, around Victoria Harbor. Thus high concentrations of air pollutions in side the street canyons were expected in the region. The climate in Hong Kong is sub-tropical with four seasons, under influence of the Asian monsoon. In winter, northeasterly wind becomes the most prevailing wind. Continental emissions from the interior Asia intrude into Hong Kong and the South China Sea with northeasterly wind. Pollution episodes observed by EPD (http://www.epd.gov.hk/) were thought to associate with stagnating high-pressure systems that brought subsiding polluted airstreams over Hong Kong, transported from north or northeast (South China). Therefore the levels of air quality in Hong Kong are universally high in winter and transit period of spring due to the influence not only from local sources but also from polluted air mass transported from South China.

At present, Hong Kong Environment Protection Department (HKEPD) has recognized the two air pollution issues faced (http://www.epd.gov.hk/). One is local street-level pollution. The other is the regional smog problem. Diesel vehicles are the main source of street-level pollution. Smog, however, is caused by a combination of pollutants from motor vehicles, industry and power plants both in Hong Kong and in the Pearl River Delta region. Although several studies on PM_{10} (Yu et al. 2004; Qin et al. 1997) and $PM_{2.5}$ (Louie et al. 2004; Ho et al. 2003; Cao et al. 2003) in Hong Kong have been conducted in the past, the studies that focus on the roadside pollution and regional pollution are quite limited and few studies has been given to investigation on $PM_{1.0}$, which is quite significant because of it is dominated proportion in vehicle exhausts and in air masses transported from remote area. Therefore continuous 24-hour measurements on $PM_{1.0}$ and $PM_{2.5}$ were launched near a major road from January 21 to May 31, 2004. For the purpose of comparison, $PM_{2.5}$ mass data from four monitoring stations operated by EPD was also reported. One of them is ground-level roadside monitoring station (Central) and the other three are general ambient monitoring stations (Tap Mun, Tsuan Wan, and Tung Chung). In this study, winter was classified as those days before March 1st, and spring was the days after March 1st. The objectives of this study are: to determine the levels of fine particles ($PM_{1.0}$ and $PM_{2.5}$) and carbonaceous aerosols in roadside environment; to explore the relationship of mass, OC, and EC between $PM_{1.0}$ and $PM_{2.5}$ in roadside environment; to characterize the meteorological parameter and chemical properties of fine particles during the highest PM days (episode days) and the lowest PM days (days that local vehicle emissions were dominated source); to identify the factors that induce the significant fluctuations of fine particles and carbonaceous aerosols.

2. METHODOLOGY

Sampling Site

The measurements of $PM_{1.0}$ (fine particle with diameter less than 1 μm) and $PM_{2.5}$ (fine particle with diameter less than 2.5 μm) were took place at the roadside air quality monitoring site in the campus of the Hong Kong Polytechnic University (PU) (Figure 1) that locates near Victoria Harbor. It is the most residential and commercial areas in Hong Kong around Victoria Harbor. The sampling was conducted from January 21, 2004 to May 31, 2004, yielding a total of 132 days of data. The sampling location, PU roadside station, is about one meter away from the curb of Hong Chong Road, which leads to the Cross Harbor Tunnel (one of the two busiest cross-harbor tunnels in Hong Kong, having four lanes to each direction), with the objective to investigate air quality experienced by the pedestrians in an environment close to vehicular traffic. The traffic flow on Hong Chong Road is extremely high, with about 119,759 per day (the 2003 annual traffic census). No industries or other anthropogenic sources were found in the vicinity. Thus the PU roadside was considered to be a typical roadside environment in Hong Kong, with significant influence from primary vehicular emissions. The presented traffic data were obtained in terms of the number of cars running in both directions per day.

Sample Collection

Daily $PM_{1.0}$ and $PM_{2.5}$ samples were collected with two-paralleled Plus Model 2025 Sequential Air Sampler (Rupprecht and Patashnick Co. Inc., Albany, NY) operated at 16.7 l min^{-1}. The particles were collected on quartz filters with 47 mm in diameter, which were weighted twice before and after sampling using a Microbalance (Sartorius Microbalance, Sartorius AG, Goettingen, Germany) with the sensitivity of 1 μg in the 0-250 mg range.

Before sampling, quartz filters were preheated in an electric furnace at 800℃for at least 3 hours in order to remove carbonaceous contaminants. The samples were balanced in a desiccator with temperature of 20℃-30 ℃ and relative humidity of 30%-40% for 24 hours before and after sampling. The filters are handled only with tweezers cleaned by dry KimWipes (Kimberly-Clark Corporation, USA) to reduce the possible of contamination. After weighting, sampled filters were stored in a refrigerator at about 4℃ 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 Desert Research Institute (DRI) Thermal/Optical Carbon Analyzer Model 2001 with the IMPROVE thermal/optical reflectance (TOR) protocol (Chow et al. 1993). The DRI thermal/optical carbon analyzer is based on the preferential oxidation of organic carbon (OC) and elemental carbon (EC) compounds at different temperatures. It relies on the fact that organic compounds can be volatilized from the sample deposit in a helium (He) atmosphere at low temperatures, while elemental carbon is not oxidized and removed. The TOR protocol heats the sample on quartz filter with a punched area of 0.526 cm^2 stepwise

at temperatures of 120□ (OC1), 250□ (OC2), 450□ (OC3) and 550□ (OC4) in a non-oxidizing helium atmosphere, and 550□ (EC1), 700□ (EC2) and 800□ (EC3) in an oxidizing atmosphere with 2% oxygen in helium. The carbon evolved is oxidized to carbon oxide (CO_2) and then reduced to methane (CH_4) for quantification with a flame ionization detector (FID). The pyrolysis of OC is continuously monitored by a 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, whereas the carbon evolved beyond this temperature is defined as EC. The minimum detection limit (MDL) of carbon combustion methods is 0.82 μg cm^{-2} for total organic carbon, 0.19 μg cm^{-2} for total elemental carbon, and 0.93 μg cm^{-2} for total carbon. All of samples in this study have concentrations higher than MDL. Replicated analyses were performed for ~10% of all samples.

$PM_{2.5}$ Monitoring Sites and Data Collection

The air quality monitoring network in Hong Kong operated by the HKEPD comprises fourteen fixed monitoring stations. At the three general stations (Tung Chung, Tsuan Wan, and Tap Mun) and one roadside station (Central), $PM_{2.5}$ mass were routinely monitored with the Tapered Element Oscillating Microbalance (TEOM). Figure 1 shows the geographical distribution of stations. The ambient stations located on rooftops of four- to six-storey buildings, with unobstructed air flows from most directions. The selection of these three monitoring stations was to evaluate the air quality in ambient atmosphere. Besides, the three monitoring stations cross through the whole Hong Kong territory from northeast to southwest, thus providing a good spatial representation of air quality in Hong Kong. The data of roadside station (Central) was used to make comparison with the results of PU roadside station in this study.

3. DISCUSSION

Concentrations of Mass, OC and EC

Table 1 summarized the average mass concentrations of $PM_{1.0}$ and $PM_{2.5}$ measured at PU roadside station. On average, the mass concentrations of $PM_{1.0}$ and $PM_{2.5}$ were 35.9±12.4 μg m^{-3} and 52.3±18.3 μg m^{-3}, respectively, throughout the total 132 sampling days. The ranges of particle concentrations varied significantly, varying from 17.5 μg m^{-3} to 85.0 μg m^{-3} for $PM_{1.0}$ and from 24.3 μg m^{-3} to 111.4 μg m^{-3} for $PM_{2.5}$, respectively. Observed $PM_{2.5}$ loading at PU roadside were comparable with Central (the average concentration in Central is 53.3±21.7 μg m^{-3}), another roadside station operated by HKEPD. This gives confidence that the measurements at PU station represent a typical roadside level. The $PM_{2.5}$ mass showed a clear spatial pattern in Hong Kong (Table 1). The average mass concentrations of $PM_{2.5}$ collected from PU roadside and Central roadside were higher than the other three ambient monitoring stations (Tung Chung, Tsuan Wan, and Tap Mun). Among the ambient stations, high $PM_{2.5}$ levels were observed in urban atmosphere (43.2±24.1 μg m^{-3} in Tung Chung and 43.4±21.6 μg m^{-3} in Tsuan Wan), while in background atmosphere, the lowest particle loading were obtained (35.9±16.2 μg m^{-3} in Tap Mun). Tap Mun had the lowest average mass concentration because it located on a small island, surrounding by marine. No local pollution sources exist in the vicinity. All of mass

data measured at ambient stations showed a clear seasonality with high concentrations in winter and low concentrations in summer (Table 1). While this trend was ambiguous in roadside environment (Table 1).

A statistical summary of the daily average concentration of OC and EC in $PM_{1.0}$ at PU roadside are given in Table 1. The overall average concentrations of OC and EC were 7.0±2.6 μg m^{-3} and 9.5±3.5 μg m^{-3}, respectively. OC showed a higher level in winter (8.3±3.3 μg m^{-3}) than in spring (6.4±1.9 μg m^{-3}), while EC was slightly higher in spring (9.7±3.3 μg m^{-3}) than in winter (9.0±3.8 μg m^{-3}). Up to now, the data of carbonaceous content in $PM_{1.0}$ was scarce throughout the world. Li and Lin (2002) measured the TC and EC concentrations of $PM_{1.0}$ via a combustion technique (Table 2), which was conducted in Taipei during October of 1999. The concentrations of OC (11.5±2.8 μg m^{-3}) and EC (11.3±1.7 μg m^{-3}) observed at the traffic station were slightly higher than that in this study. While the levels of carbonaceous aerosols collected in this study were much higher than that obtained in urban ambient atmosphere of Taipei (3.4±0.9 μg m^{-3} and1.3±0.7 μg m^{-3} for $PM_{1.0}$ and $PM_{2.5}$, respectively).

The daily average concentration of OC in $PM_{2.5}$ was 11.0±4.7 μg m^{-3} with a higher concentration in winter (54.1±21.1 μg m^{-3}) and lower concentration in spring (51.1±17.0 μg m^{-3}). While the average concentration of EC was 12.2±4.4 μg m^{-3} with a higher value (12.7±4.2 μg m^{-3}) in spring than in winter (11.0±4.7 μg m^{-3}). The levels of carbonaceous aerosols were compared with previous studies listed in table 2. Since there is no standard analysis method till now, different thermal methods were used in those studies, which might result in large difference in OC and EC measurements. The OC and EC measured in this study were a little higher than those observed in previous studies (Cao et al. 2003; Ho et al. 2003) (Table 2). This is due to the different distance between this study (1 meter away) and previous studies (more than 30 meters away). Zhu et al. (2002) observed dramatic decrease of number concentrations of ultrafine particles near the 710 freeway when measurement moved from 4 meters away from the road curb to 17 meters away. This suggested that the increased carbonaceous levels in this study mainly attribute to much closer distant from road. The OC concentration in $PM_{2.5}$ was comparable to the results from other roadside studies in Hong Kong (Yu et al. 2004) and in Macao (Wu et al. 2003). However, the EC concentration observed at the roadsides of Hong Kong (this study; Yu et al. 2004) was higher than Macao. This perhaps due to the higher diesel vehicle fractions in Hong Kong since the EC abundance in diesel-fueled vehicles could be twice that of gasoline-fueled vehicles (Watson et al. 1994). Generally speaking, both of OC and EC concentrations in $PM_{2.5}$ measured at a roadside in Hong Kong were much higher than those in urban atmosphere (Cao et al. 2003; Wu et al. 2003; Lin and Tai 2001; Lee and Kang 2001; Ohta et al.1998).

On average, $PM_{1.0}$ consisted 19.4% of OC and 26.4% of EC, respectively, and $PM_{2.5}$ consisted 21.1% of OC and 23.4% of EC. The concentrations of mass, OC, and EC in $PM_{1.0}$ and $PM_{2.5}$ were quite similar between weekdays and Saturday, while slightly reduced concentrations were observed on Sunday due to the decreased of vehicular traffic.

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

Particles with different size ranges possibly differ not only in size and morphology but also in formation mechanisms; sources; and chemical, physical, and biological properties. Several studies have been conducted to explore the relationship between $PM_{1.0}$ and $PM_{2.5}$ mass. Cabada et al. (2004) found that there were good correlations existed between $PM_{2.5}$ and PM_x（particles with diameter less than x μm）, when PM_x is close to $PM_{2.5}$, through investigating the relationship among different particle fractions in Pittsburgh Supersite with MOUDI sampler. Vallius et al. (2000) and Keywood et al. (1999) reported the same findings and suggested that there were similar sources between $PM_{1.0}$ and $PM_{2.5}$. In contrast, Lundgren et al. (1996) evaluated the $PM_{1.0}/PM_{2.5}/PM_{10}$ characteristics in Phoenix, Arizona with a trichotomous sampler. It was concluded that $PM_{1.0-2.5}$ was caused by dispersion aerosol, while $PM_{1.0}$ minimized interference from natural sources compared with $PM_{2.5}$ and PM_{10}. Therefore $PM_{1.0}$ was considered to be a better indicator of primary particles in a roadside microenvironment than $PM_{2.5}$.

The relationship between $PM_{1.0}$ and $PM_{2.5}$ is still controversial up to now. Regression analysis was conducted in two seasons in this study, as illustrated in Figure 2. In winter, very good correlations (r=0.97) exist between $PM_{1.0}$ and $PM_{2.5}$ mass. On average, around 70% of the $PM_{2.5}$ mass was in particles smaller than 1 μm. While the correlation is moderate (r=0.80) in spring and $PM_{1.0}$ contributed to only about 50% of $PM_{2.5}$. The regression analyses for OC in $PM_{1.0}$ and $PM_{2.5}$ reported the similar relationship for winter and spring, while the correlation coefficient in spring (r=0.78) is much lower than that in winter (r=0.91). OC in $PM_{1.0}$ accounted for 50-60% of OC in $PM_{2.5}$. This result demonstrated that the relationship between $PM_{1.0}$ and $PM_{2.5}$ varied in terms of different seasons. Factors related meteorological conditions perhaps contributed to the changes of relationship between $PM_{1.0}$ and $PM_{2.5}$. However, good correlation between $PM_{1.0}$ and $PM_{2.5}$ was measured for EC concentrations in both winter (r=0.96) and spring (r=0.88). $PM_{1.0}$ EC accounted for 70-80% of $PM_{2.5}$ EC. Generally, EC originates from incomplete combustion of carbon-containing material and does not formed in atmosphere due to its nearly inert property (Ogren and Charlson, 1983). The main source of EC in urban atmosphere is relative simple, mostly from vehicular emissions in this study. This perhaps explained the reason of good correlation of EC between $PM_{1.0}$ and $PM_{2.5}$ regardless of seasonal difference.

Time Series of Fine Particles

Figure 3 shows the daily variability of $PM_{1.0}$ and $PM_{2.5}$ at PU roadside station from January 2004 to May 2004. The trend was quite similar between $PM_{1.0}$ and $PM_{2.5}$ with a high correlation coefficient (R^2) of 0.86. Time series of fine particles exhibited seven episode days in January 30, February 14-15, February 23, February 26, April 19-20, respectively, with higher $PM_{1.0}$ and $PM_{2.5}$ levels than the observed average concentration (35.9±12.4μg m^{-3} and 52.3±18.3 μg m^{-3} for $PM_{1.0}$ and $PM_{2.5}$, respectively) by a factor of two (Table 3). These days had the highest 5% PM during the whole measuring days. Central also showed the same peaks in the same episode days and the average loading of $PM_{2.5}$ was two times higher than the average concentration (53.3±21.7 μg m^{-3}). The same phenomena were also observed at

ambient monitoring stations (Tung Chung, Tsuan Wan, Tap Mun) during the same episode days. For instance, the average $PM_{2.5}$ loading exceeded the average level (43.4±21.6 μg m^{-3}) during the same high aerosol days by a factor of 2.4 at Tsuan Wan (Figure 3). This suggested that the observed episode days with about two times higher PM loading at PU roadside reasonably represent fine particle episodes in a regional scale.

Of the seven episode days, five days in winter were dominated by northeasterly winds (wind direction 0°–90°) and all of daily wind speed exceeded 15 km h^{-1} with maximum of 42.7 km h^{-1}, except for February 14 (8.7 km h^{-1}). This suggested air mass originated from northeast region might be a major factor to observed high level of fine particles in most episode days. While in the other two days of April there were a high-pressure ridge observed over southeast China. Therefore although the prevailing wind was from the south (wind direction 90°–270°), namely from marine, fine particles still accumulated due to the subsiding airstreams and relative low mixing height.

Carbonaceous aerosols showed quite similar fluctuations with fine particles observed at PU roadside (Figure 4), especially for OC. Figure 4 only displays the time series of mass, carbonaceous aerosols in $PM_{1.0}$. The OC concentrations in $PM_{1.0}$ and $PM_{2.5}$ were about two times higher than observed average concentrations (7.0 μg m^{-3} and 11.0 μg m^{-3} for $PM_{1.0}$ and $PM_{2.5}$, respectively) during episode days, and EC increased by a factor of 1.3 for both $PM_{1.0}$ and $PM_{2.5}$, reflecting carbonaceous aerosols were affected in a certain degree by regional pollution episodes although vehicular emissions were believed to be the main source at PU roadside. While, during episode days, TC only accounted for 33.4% and 37.3% of $PM_{1.0}$ and $PM_{2.5}$ (Table 3), which are much lower than average contributions (about 47.5% and 46.4% for $PM_{1.0}$ and $PM_{2.5}$, respectively). This suggested that although elevated concentrations existed for carbonaceous aerosols during episode days, non-carbonaceous aerosols occupied a large number of proportions of $PM_{1.0}$ and $PM_{2.5}$. The above calculation about carbonaceous aerosols excluded February 14, in which wind speed is the third lowest (8.7 Km h^{-1}, Table 3) during measuring days of winter (average wind speed in winter was 23.5 Km h^{-1}). The concentrations of OC and EC in $PM_{1.0}$ (23.3 μg m^{-3} and 22.9 μg m^{-3}, respectively) and $PM_{2.5}$ (35.8 μg m^{-3} and 28.6 μg m^{-3}, respectively) were much higher than other episode days (Table 3) and TC accounted for 54.4% of $PM_{1.0}$ and 62.5% of $PM_{2.5}$, respectively.

For the purpose of comparison, the lowest 5% $PM_{1.0}$ and $PM_{2.5}$ days, which were the same days for $PM_{1.0}$ and $PM_{2.5}$, was selected. As showed in Table 3, they were April 25, May 3, May 12-14, and May 30-31, with the average mass of 20.8 μg m^{-3} for $PM_{1.0}$ and 27.5 μg m^{-3} for $PM_{2.5}$, respectively. The wind in these days exclusively was from marine. The airstream originating from marine brought relative clean air to Hong Kong. Thus local street-level source was expected to be main contributor to air pollution. $PM_{2.5}$ on May 12-14 and May 30-31 were also within the lowest 5% $PM_{2.5}$ days in ambient (Tap Mun, Tsuan Wan, and Tung Chung). Carbonaceous aerosols were dominated species in the lowest 5% PM days at PU roadside station, with averaged 88.9% and 78.5% of TC in $PM_{1.0}$ and $PM_{2.5}$, respectively. The average

proportion of EC exceeded 50% for both $PM_{1.0}$ and $PM_{2.5}$. Average OC/EC ratios in $PM_{1.0}$ and $PM_{2.5}$ were 0.6 and 0.5, respectively, which were much lower than those observed in episode days (1.0 and 1.5 for $PM_{1.0}$ and $PM_{2.5}$, respectively).

In winter, wet scavenging effect was very clear for particle mass and OC through the comparison between rainy days (including drizzle) and non-rainy days (Table 4). The average concentrations of $PM_{1.0}$ and $PM_{2.5}$ in rainy days at PU roadside station were 32.1±2.5 μg m^{-3} and 40.8±4.7 μg m^{-3}, respectively, which is much lower than particle loading in non-rainy days, about 42.5±16.3 μg m^{-3} for $PM_{1.0}$ and 56.5±22.0 μg m^{-3} for $PM_{2.5}$. OC concentration also showed low levels in rainy days (6.9±1.1 μg m^{-3} and 9.8±3.9 μg m^{-3} in $PM_{1.0}$ and $PM_{2.5}$, respectively) and high levels in non-rainy days (8.5±3.9 μg m^{-3} and 8.9±3.9 μg m^{-3} in $PM_{1.0}$ and $PM_{2.5}$, respectively) in winter. The same phenomena were observed in ambient atmosphere (Table 4). Although rainfall is much heavier in spring than in winter, it seemed that the relationship between particles and precipitation was not very clear (Table 4). As mentioned above, Hong Kong was influenced by not only local source but also regional source in winter. Elevated OC concentrations observed in winter maybe were due to the contribution of secondary organic matter in transported aged air mass (Yu et al. 2004). Rainout was thought to be the principal removal mechanism (Pandis et al.1993) for aged aerosol since the presence of more soluble secondary aerosols (e. g. organic materials and sulfates). However, in spring less secondary aerosols present in atmosphere, which leads to the low wet removal rate.

Although wet scavenging is likely to be the dominant removal mechanism for EC (Ogren and Charlson, 1983), the effect is believed to depend on the extent to which the EC is coated with hygroscopic substance (Ogren et al. 1984). In other words, it depends on whether EC is internal mixture or external mixture. An internal mixture is one where EC is physically in contact with other, perhaps more abundant, hygroscopic aerosol constituents (e.g., sulfates), with the other compounds dominating the physical and chemical properties of the EC-containing particles (Ogren and Charlson, 1983). External mixture defines EC physically isolated from other constituents, such that the properties of the EC-containing particles with respect to water are determined by EC alone (Ogren and Charlson, 1983). Since urban aerosol samples taken near a source region were reported to exhibit a large degree of external mixing state (Covert and Heintzenberg, 1984). Therefore it is not surprised the poor wet scavenging effect of EC observed (Table 4).

Human behavior is another factor affecting the PM mass concentration and it's carbonaceous proportion. It was public holiday for Chinese Lunar New Year from January 22 to 25 in Hong Kong, in which most of commercial activities and production activities stopped. PM mass concentrations decreased gradually due to less passing traffic on the road, especially for diesel-fueled vehicles. On January 25, the lowest mass concentration in winter observed for both $PM_{1.0}$ and $PM_{2.5}$. The average OC in $PM_{1.0}$ during Chinese Lunar New Year accounted for 23.4% of $PM_{1.0}$ mass, which was higher than observed average value (19.7%) throughout the whole measuring period. While EC only accounted for 19.4% of $PM_{1.0}$, which was lower than observed average value (27.8%).

4. CONCLUSION

Aerosol fine particle samples collected continuously at the heavily-traffic road in Hong Kong, PU roadside station, provided a near-five-month data set in winter and spring. An integrated data set, including aerosol mass, the concentration of carbonaceous aerosols associated episode and non-episode days, has been obtained, which is especially important because it can virtually help us enhance the understand of two important air pollution problems faced by Hong Kong: the local-street pollution and regional smog pollution.

Continuous measurements of $PM_{1.0}$ and $PM_{2.5}$ over nearly 5 months in PU roadside showed that carbonaceous aerosols were major components in fine particles, constituting 45.7% of $PM_{1.0}$ (35.9±12.4 μg m^{-3}) and 44.4% of $PM_{2.5}$ (52.3±18.3 μg m^{-3}). The average organic carbons in $PM_{1.0}$ and $PM_{2.5}$ were 7.0±2.6 μg m^{-3} and 11.0±4.7 μg m^{-3}, respectively and the average EC concentrations were 9.5±3.5 μg m^{-3} and 12.2±4.4 μg m^{-3}, respectively. Particle mass and OC showed a slightly higher concentrates in winter than that in spring. The $PM_{2.5}$ mass showed a clear spatial variability. In terms of mass concentration levels and assigning the sites in descending order with site characteristics, it is observed that roadside>urban>background.

The occurrences of episodes were observed to be commonly associated with high northeasterly wind in winter, while low wind speed from south in spring. The mean concentrations of fine particles during episode days were two times higher than the average value throughout the whole measuring days. Although elevated concentrations of OC and EC were observed during episode days, the contributions of carbonaceous aerosols were extremely lower than that in the lowest PM days. For example, the fraction of $PM_{1.0}$ accounted for by OC and EC was ~16% and ~17%. While PM1.0 was composed of ~22% for OC and ~49% for EC, respectively, when a local-street pollution was dominate source for particle aerosols.

In winter, rainout might be the principal removal mechanism for mass and OC since the presence of soluble secondary aerosols (e. g. organic materials and sulfates) in transported aged air mass. While no removal effect was found for EC. In addition, less commercial activities and human activities also lead to lower particle levels.

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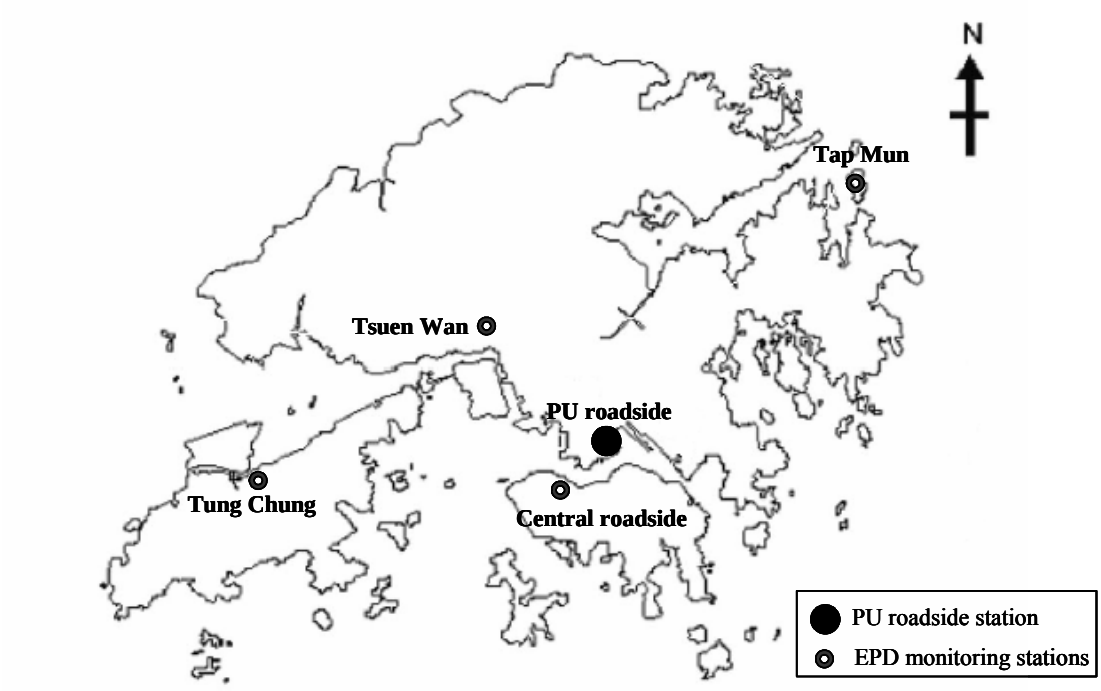

Figure 1. Sampling locations

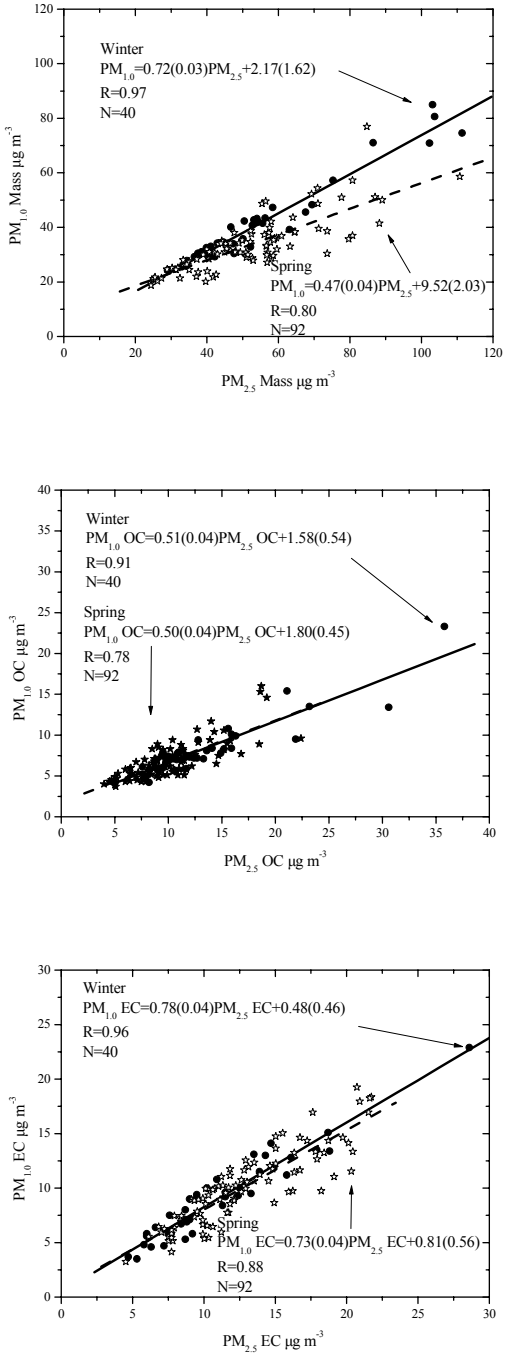

Figure 2. Correlations of Mass, OC, and EC between $PM_{1.0}$ and $PM_{2.5}$. Standard error is in bracket.

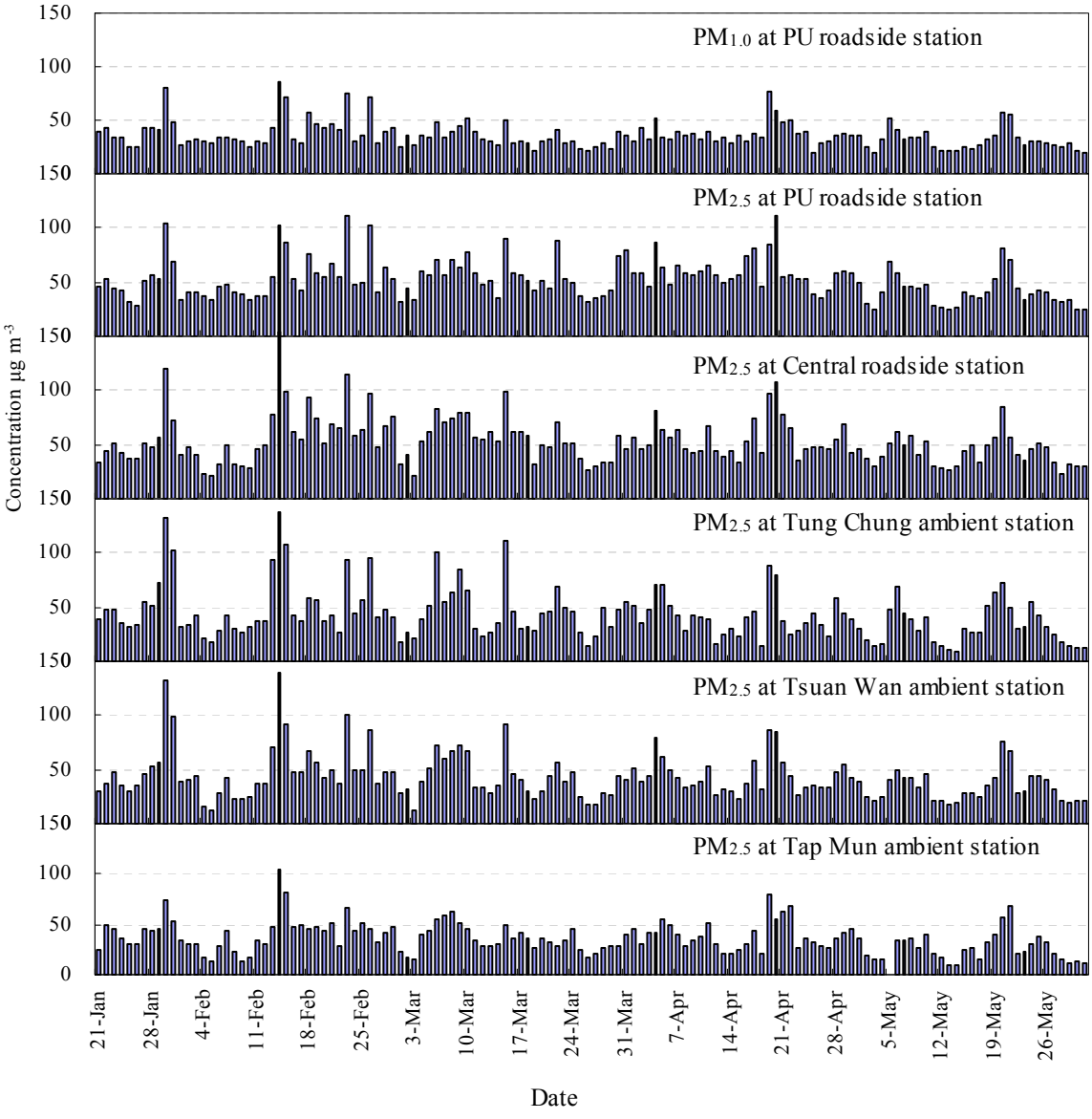

Figure 3. Daily 24-h average $PM_{1.0}$ and $PM_{2.5}$ concentrations at PU roadside and monitoring stations operated by EPD during January to May 2004.

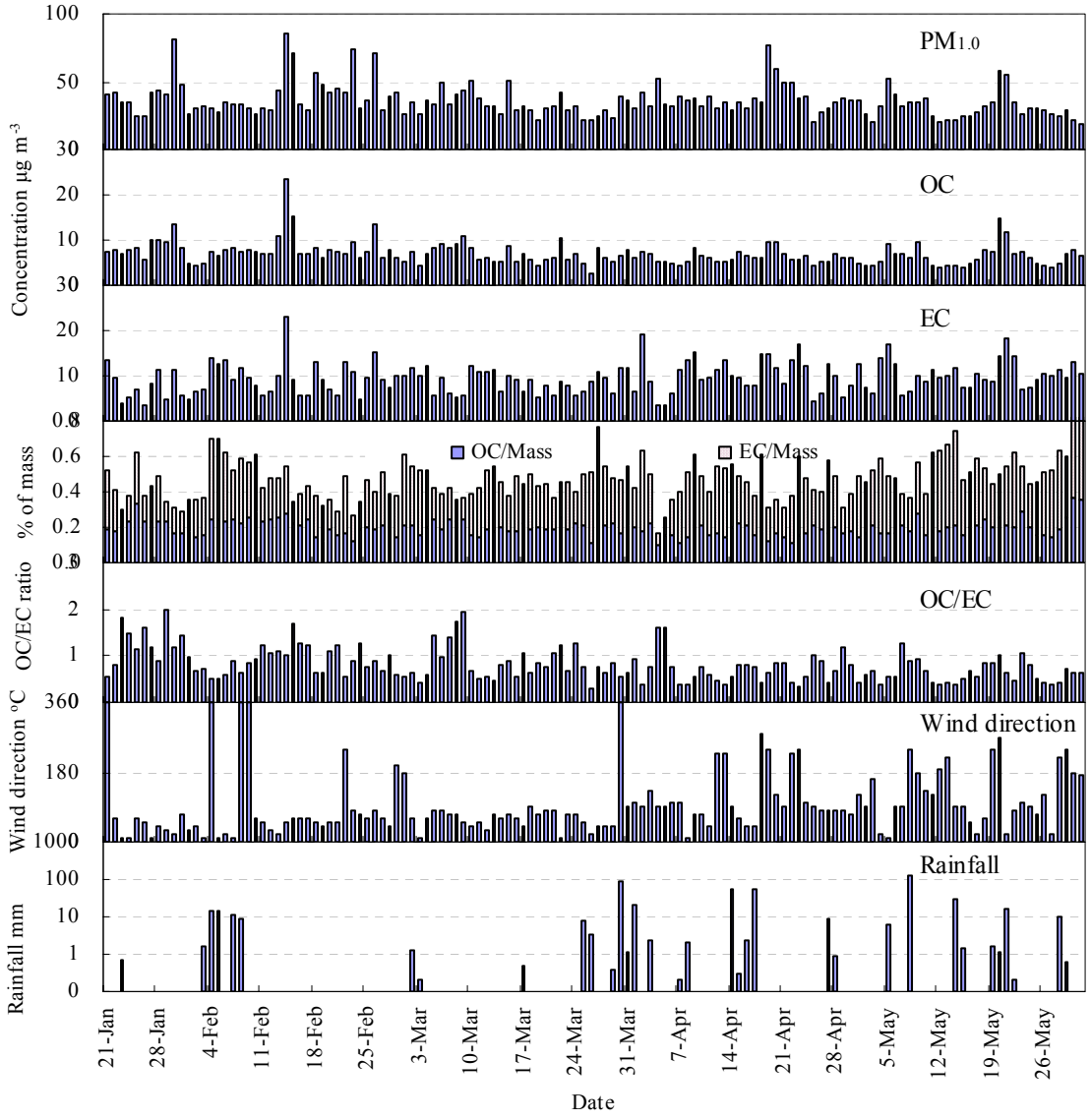

Figure 4. Daily 24-h average $PM_{1.0}$, OC, EC concentrations, and OC/mass, EC/mass, OC/EC ratios at PU roadside during January to May 2004, with corresponding meteorological data.

Table 1. Seasonal average of particle mass and carbonaceous aerosols in PU roadside station and four monitoring stations

		PU roadside						Central[a]	Tung Chung[b]	Tsuan Wan[b]	Tap Mun[b]
		$PM_{1.0}$ ($\mu g\ m^{-3}$)			$PM_{2.5}$ ($\mu g\ m^{-3}$)			$PM_{2.5}$ ($\mu g\ m^{-3}$)			
		Mass	OC	EC	Mass	OC	EC	Mass	Mass	Mass	Mass
Winter	Jan	41.4± 15.1	8.6± 2.1	7.7± 3.4	52.7± 20.3	13.9± 3.9	10.3± 4.4	54.1± 24.3	58.8± 31.7	55.0± 32.1	43.7± 13.6
	Feb	40.8± 15.9	8.2± 3.7	9.6± 3.9	54.6± 21.7	13.0± 6.6	11.3± 4.9	61.1± 30.4	49.4± 28.5	49.6± 27.3	41.3± 19.4
	Average	40.9± 15.5	8.3± 3.3	9.0± 3.8	54.1± 21.1	13.2± 6.0	11.0± 4.7	59.2± 28.7	52.0± 29.3	51.1± 28.4	42.0± 17.9
Spring	Mar	33.1± 8.1	6.7± 1.9	8.6± 2.4	54.2± 16.3	11.0± 3.2	11.3± 3.1	53.8± 18.6	44.8± 23.3	41.5± 18.6	35.1± 12.1
	Apr	37.2± 11.4	6.1± 1.3	10.0± 4.1	59.6± 15.4	11.0± 3.6	14.7± 4.8	56.0± 17.5	41.5± 17.8	44.9± 16.7	39.5± 14.4
	May	31.0± 9.7	6.4± 2.4	10.4± 3.1	40.9± 14.0	8.2± 3.6	12.3± 3.9	42.6± 13.3	31.8± 17.9	33.8± 14.0	25.6± 14.3
	Average	33.8± 10.1	6.4± 1.9	9.7± 3.3	51.5± 17.0	10.0± 3.7	12.7± 4.2	50.8± 17.4	39.4± 20.4	40.0± 17.0	33.3± 14.7
Total		35.9± 12.4	7.0± 2.6	9.5± 3.5	52.3± 18.3	11.0± 4.7	12.2± 4.4	53.3± 21.7	43.2± 24.1	43.4± 21.6	35.9± 16.2

[a] Roadside monitoring station; b Ambient monitoring station

Table 2. Levels of carbonaceous aerosols in urban atmosphere in Asia.

Location	Sampling Period	Size	TC	OC	EC	OC/EC	Location	Method	Reference
			($\mu g\ m^{-3}$)	($\mu g\ m^{-3}$)	($\mu g\ m^{-3}$)				
Hong Kong, PU	2004.1-2	$PM_{1.0}$	17.3	8.3 ± 3.3	9.0 ± 3.8	1	Roadside	IMPROVE-TOR	This study
Tai Wan, Taipei	1999.1	$PM_{1.0}$	22.9	11.5 ± 2.8	11.3 ± 1.7		Roadside	Combustion	Li C.S. 2002
Tai Wan, Taipei	1999.12	$PM_{1.0}$	4.7	3.4 ± 0.9	1.3 ± 0.7		Urban	Combustion	Li C.S. 2002
Hong Kong, PU	2004.1-2	$PM_{2.5}$	24.2	13.2 ± 6.0	11.0 ± 4.7	1.3	Roadside	IMPROVE-TOR	This study
Hong Kong, PU	2000.11-2001.2	$PM_{2.5}$	15.3	9.5 ± 2.0	5.8 ± 1.1	1.6	Roadside	TMO[a]-EA[b]	Ho K.F. 2003
Hong Kong, PU	2002.1-2	$PM_{2.5}$	16.7	10.6 ± 3.7	6.1 ± 1.8	1.7	Roadside	IMPROVE-TOR	Cao J.J. 2003
Hong Kong, MK	2000.11-2001.10	$PM_{2.5}$	36.8	16.7 ± 7.6	20.2 ± 4.2	0.8	Roadside	IMPROVE-TOR	Louie P. 2004
Hong Kong, TW	2000.11-2001.10	$PM_{2.5}$	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	$PM_{2.5}$	5.9	4.2 ± 3.7	1.7 ± 0.9	2.5	Rural	IMPROVE-TOR	Louie P. 2004
Tai Wan, Taipei	1999.1	$PM_{2.5}$	23.6	12.2 ± 3.0	11.4 ± 2.5		Roadside	Combustion	Li C.S. 2002
Tai Wan, Taipei	1999.12	$PM_{2.5}$	6.2	3.4 ± 0.5	2.9 ± 0.3		Urban	Combustion	Li C.S. 2002
China, Guang Zhou	2002.1-2	$PM_{2.5}$	23.8	17.8 ± 10.2	6.0 ± 3.2	2.9	Urban	IMPROVE-TOR	Cao J.J. 2003
China, Shen Zhen	2002.1-2	$PM_{2.5}$	19.2	13.2 ± 4.1	6.1 ± 1.8	2.2	Urban	IMPROVE-TOR	Cao J.J. 2003
China, Zhu Hai	2002.1-2	$PM_{2.5}$	17.3	12.2 ± 4.4	5.0 ± 1.6	2.4	Urban	IMPROVE-TOR	Cao J.J. 2003
China, Macao	2001.11-12	$PM_{2.5}$	16.6	12.2	4.4	2.8	Roadside	TOT-EA[b]	Wu Y. 2003
Tai Wan	1998.11 - 1999.4	$PM_{2.5}$	14.5	10.4	4	2.6	Urban	EA[b]	Lin J. J. 2001
Korea, Chongju	1995, Fall	$PM_{2.5}$	12.4	6	6.4	0.9	Urban	IMPROVE-TOR	Lee H. S. 2001
	1995, Winter	$PM_{2.5}$	9.3	5	4.3	1.2	Urban	IMPROVE-TOR	Lee H. S. 2001
Japan, Sapporo	1992.9-10	$PM_{2.0}$	9.1	4.1	5	0.8	Urban	EA[b]	Ohta S. 1998
	1992.1-2	$PM_{2.0}$	9	3.9	5.1	0.8	Urban	EA[b]	Ohta S. 1998
Hong Kong, MK	2001.1-12	PM_{10}	26.8	16.3 ± 6.2	10.5 ± 2.9	1.6	Roadside	NOISH-TOR	Yu J.Z. 2004
Japan,Uji	1998.9-10	PM_{10}	6.4	1.8	4.6	0.4	Urban	R&P 5400	Holler R. 2002
	1998.11-12	PM_{10}	10.2	2.5	7.7	0.3	Urban	R&P 5400	Holler R. 2002

[a] Thermal manganese dioxide oxidation

[b] Elemental analysis

Table 3. Summary statistics for the concentrations of PM, carbonaceous aerosols; and ratios during the highest and lowest 5% PM days in 2004 with corresponding meteorological data.

			The highest 5% PM days (episode days)							The lowest 5% PM days						
Date			30 Jan	14 Feb	15 Feb	23 Feb	26 Feb	19 Apr	20 Apr	25 Apr	3 May	12 May	13 May	14 May	30 May	31 May
$PM_{1.0}$	Mass	$\mu g\ m^{-3}$	80.6	85.0	71.0	74.6	70.9	77.3	59.0	20.5	20.1	20.7	21.5	21.8	22.1	19.1
	OC	$\mu g\ m^{-3}$	13.5	23.3	15.4	9.5	13.4	9.4	9.6	4.3	4.3	3.7	4.3	4.5	8.0	6.7
	EC	$\mu g\ m^{-3}$	11.5	22.9	9.2	10.8	15.1	14.8	11.6	4.2	6.3	9.4	10.0	11.7	13.0	10.3
	OC/EC		1.2	1.0	1.7	0.9	0.9	0.6	0.8	1.0	0.7	0.4	0.4	0.4	0.6	0.6
	$OC/PM_{1.0}$	%	16.7	27.4	21.7	12.7	18.9	12.2	16.3	21.0	21.4	17.9	20.0	20.6	36.2	35.0
	$EC/PM_{1.0}$	%	14.3	26.9	13.0	14.5	21.3	19.1	19.7	20.5	31.3	45.4	46.5	53.7	59.0	53.9
$PM_{2.5}$	Mass	$\mu g\ m^{-3}$	103.7	103.1	86.5	111.4	102.3	84.8	110.8	39.7	24.3	26.2	25.1	27.3	25.5	24.4
	OC	$\mu g\ m^{-3}$	23.2	35.8	21.1	21.9	30.6	13.9	22.4	6.0	4.5	5.1	4.5	5.0	8.1	7.0
	EC	$\mu g\ m^{-3}$	13.9	28.6	11.5	10.9	18.7	15.0	20.3	7.7	7.2	10.3	10.1	12.8	15.5	12.2
	OC/EC		1.7	1.3	1.8	2.0	1.6	0.9	1.1	0.8	0.6	0.5	0.4	0.4	0.5	0.6
	$OC/PM_{2.5}$	%	22.4	34.7	24.4	19.7	30.0	16.3	20.2	15.2	18.5	19.2	17.9	18.3	31.7	28.5
	$EC/PM_{2.5}$	%	13.4	27.8	13.3	9.7	18.3	17.6	18.3	19.5	29.6	39.4	40.2	46.7	60.8	50.0
	RH	%	83	59	66	72	80	69	79	88	80	81	80	90	81	76
	Rainfall	mm												31.6		
	WD[a]	^oC	20	50	60	80	80	240	120	90	160	190	220	90	180	170
	WS[b]	$Km\ h^{-1}$	15.1	8.7	28.9	42.7	18.2	7.5	6.1	31.5	10.8	10.7	12.8	22.8	14.4	7.5

[a] Wind direction, [b] Wind speed

Table 4. Comparison of fine particles and carbonaceous aerosols between rainy days and non-rainy days.

	Season		Winter		Spring	
			Rainny days	Non-rainny days	Rainny days	Non-rainny days
	N[a]		6	34	31	61
PU roadside station	$PM_{1.0}$	$\mu g\ m^{-3}$	32.1± 2.5	42.5± 16.3	32.4± 9.3	34.4± 10.4
	$PM_{2.5}$	$\mu g\ m^{-3}$	40.8± 4.7	56.5± 22.0	52.0± 16.0	51.2± 17.6
	OC in $PM_{1.0}$	$\mu g\ m^{-3}$	6.9± 1.1	8.5± 3.5	6.4± 2.3	6.4± 1.7
	EC in $PM_{1.0}$	$\mu g\ m^{-3}$	9.8± 3.9	8.9± 3.9	10.0± 3.3	9.5± 3.4
	OC in $PM_{2.5}$	$\mu g\ m^{-3}$	10.4± 2.3	13.7± 6.3	10.5± 3.6	9.8± 3.7
	EC in $PM_{2.5}$	$\mu g\ m^{-3}$	11.9± 4.4	10.9± 4.8	13.7± 4.3	12.3± 4.1
EPD monitoring station	Central	$\mu g\ m^{-3}$	36.6± 13.2	63.2± 29.0	46.4± 14.8	53.0± 18.4
($PM_{2.5}$)	Tung Chung	$\mu g\ m^{-3}$	33.2± 12.3	55.3± 30.3	36.5± 15.7	40.8± 22.4
	Tsuan Wan	$\mu g\ m^{-3}$	30.8± 15.8	54.6± 28.8	36.2± 14.7	41.9± 17.9
	Tap Mun	$\mu g\ m^{-3}$	28.9± 13.5	44.3± 17.7	29.9± 14.3	35.1± 14.7

[a] number of days