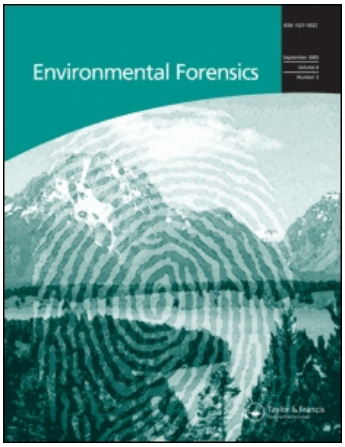

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Characteristics of Carbonaceous Aerosol at a Near–Highway-Traffic Sampling Site During Spring 2006

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The concentrations for mass, organic carbon (OC), and elemental carbon (EC) of fine ($PM_{2.5}$) and inhalable (PM_{10}) particulates were studied at a near–highway-traffic sampling site during March 6 to April 1, 2006, around central Taiwan. The primary OC/EC ratio approach is applied to assess the contribution of secondary organic aerosol (SOA) to the $PM_{2.5}$ and PM_{10} mass at the near–highway-traffic sampling site. The mass average concentrations of $PM_{2.5}$ and inhalable ( ) PM_{10} particulates were found to be 67.5 μg m^{-3} and 107.9 μg m^{-3}, respectively, during sampling period. Furthermore, analytical results indicated that the average OC and EC concentrations were 10.22 and 3.88 mg m^{-3}, respectively, in $PM_{2.5}$ particulates. The average OC and EC concentrations were 11.57 and 4.56 mg m^{-3}, respectively, in PM_{10} particulates. The average mass concentration ratio of $PM_{2.5}$to PM_{10} particulates was 1.6 during sampling period. The results also reflected that $PM_{2.5}$ particulate concentrations were the primary species (average approximately 61%). Additionally, the OC/EC ratio of both PM_{10} and $PM_{2.5}$ particulates was at all times greater than 1.7, indicating that OC constituted 66%–79% of total carbon (TC). Experimental results demonstrated that direct emissions of primary organic aerosol (POA) are greater than emissions of carbonaceous material (EC aerosol) for either PM_{10} or $PM_{2.5}$ particles. In addition, the results also reflected that OC concentration was the major species at this sampling site.

Keywords: organic carbon, elemental carbon, highway traffic, $PM_{2.5}$, PM_{10}

The carbonaceous materials are classified into two types, organic carbon (OC) and elemental carbon (EC), with respect to their chemical properties. EC and OC play important roles in various atmospheric processes. OC is a mixture of hydrocarbons and oxygenates, which can be directly emitted from sources (primary OC) or produced from atmospheric reactions involving gaseous organic precursors (secondary OC) (Pandis et al., 1992; Turpin and Huntzicker, 1995; Duan et al., 2007). Generally EC is generated by the combustion of carbon-containing fuels. EC may not only intervene in some important chemical reactions in the atmosphere (Gundel et al., 1989; Duan et al., 2007). The sources, characteristics, and potential health effects of the inhalable (diameters $<10\ \mu$m [PM_{10}]) particles, coarse ($PM_{2.5–10}$) particles (diameter in 2.5$\sim$10 μm) and fine (diameter smaller than 2.5 μm [$PM_{2.5}$]) particles () are different. $PM_{2.5}$ particulates not only readily penetrate into the lungs but also are likely to increase respiratory and mutagenic diseases (Fang et al., 2007; Cincinelli et al., 2003).

Secondary organic aerosol (SOA) contribution to $PM_{2.5}$ mass in ambient air is assessed by means of the primary OC/EC ratio approach, based on chemical data of the filters from the urban tunnel site. Organic and inorganic secondary production in the outdoor atmosphere is contributing for approximately 75% of $PM_{2.5}$ mass in winter and 40% in summer. As a consequence, effective long-term actions, as well as controlling the emissions of primary pollutants, are required for air quality standards attainment, and the potentiality of short-term interventions, such as traffic restriction, appears quite limited (Giugliano et al., 2005). Source apportionment indicated that point sources were the largest PM_{10} source at Jenwu, Linyuan, and Daliao, while at Meinung—a suburban site with less local PM_{10}, Sulfur oxides (SOx) and Nitrogen oxides (NOx) emissions—upwind boundary concentration was the major PM_{10} source, followed by point sources and top boundary concentration in southern Taiwan (Tsai and Chen, 2006). Weak correlation between OC and EC in Hong Kong can be related to the impact of the long-range transported aerosol from inland China. Average secondary OC (SOC) concentrations were

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3.8–5.9 and 10.2–12.8 μg m^{-3}, accounting for 21%–32% and 36%–42% of OC in summer and winter, respectively, in Guangzhou. Moreover, the average values of 4.2%–6.8% for SOA/ $PM_{2.5}$ indicate that SOA was minor component in $PM_{2.5}$ in Guangzhou (Duan et al., 2007).

Sampling campaigns for the production of $PM_{2.5}$ and PM_{10} filters, to be speculated for the major chemical components, have been performed at the Hungkuang University (HK) sampling site by means of a honeycomb denuder /filter pack system sampler during March 6 to April 1, 2006. Furthermore, the purpose of this study also to suggest a calculation value of the average primary OC/EC, representative of the near–highway-traffic emissions in the area around HK. The primary average OC/EC ratio approach is also applied to assess the contribution of SOA to the $PM_{2.5}$ and PM_{10} particle mass at this sampling site.

Experimental Method

Sampling Program

Figure 1 shows the sampling location for this study. Ambient particle concentration was taken on the HK campus on the roof of the Medical and Industrial Building, which is an eight-story building (25 m) and the highest sampling site on Da Du Mountain. This sampling position was near a highway and Taiwan Straits, roughly 50 m and 15 km away, respectively. The Taichung Harbor (TH) sampling site was located in the western side of central Taiwan. It occupies approximately 1540 hectares, which include 390 ha water and 1150 ha land. TH is an artificial harbor and has up to 83 ports. The sampling site for this study was located at the chemical port area, which is approximately 400 m on the east side of Taiwan Strait. The sampling height of this sampling site was approximately 10 m.

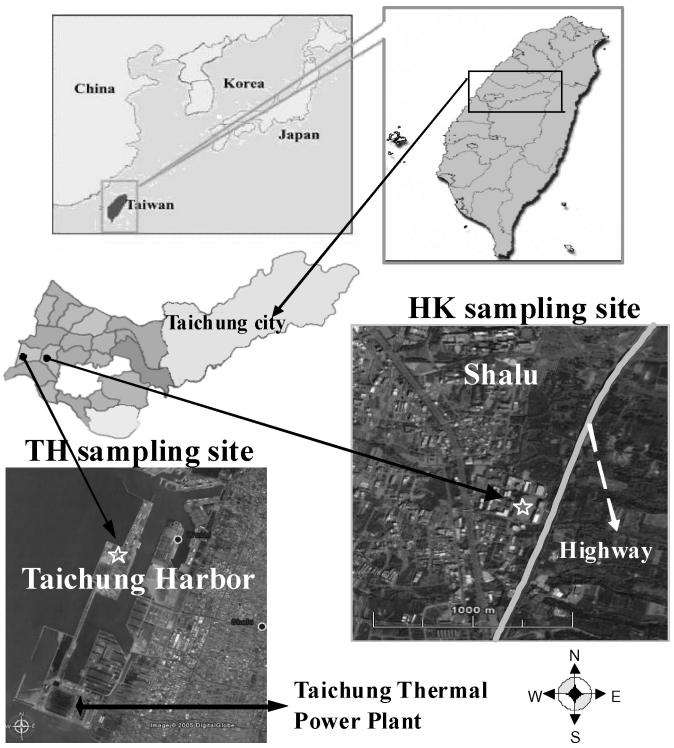


Figure 1. The location of sampling site at this highway traffic sampling site in central Taiwan. Hungkuang University (HK); Taichung Harbor (TH).

Sampling Instruments

Honeycomb Denuder.

The particulate matter ($PM_{2.5}$ and PM_{10}) were collected using the honeycomb denuder/filter pack system sampler, which had 4 channels sampling platform of particulate matter-related and gaseous species. Honeycomb denuders are small, rugged, and contain a large internal surface area. The sampling device is made up of a single cartridge that contains a well-characterized inlet with a $PM_{2.5}$ or PM_{10} impact or up to two honeycomb denuders for the removal or collection of selected gases, and a four-stage 47-mm diameter filter pack for the collection of particle-related components. $PM_{2.5}$ inlets are available for flow rates of 10 L min^{-1}, and a PM_{10} inlet is available for 10 L min^{-1}. Systems with honeycomb denuders operate at 10 L min^{-1} to maximize the collection efficiency of the denuders. The unit can also be configured without honeycomb denuders to operate as a multistage filter pack for $PM_{2.5}$ or PM_{10}. The filter pack system attached with honeycomb denuder sampler/filter pack system was applied to collect the $PM_{2.5}$ and PM_{10} only in this study.

Thermal/Optical Carbon Analysis.

The samples were analyzed for OC and EC using a DRI Model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA). The analysis procedures were modified from the method described by Cao et al. (2003). The Interagency Monitoring of Protected Visual Environments (IMPROVE) TOR precision for atmospheric samples was on the order of 5%–10% (Chow et al., 1993; 2005). This also demonstrates a reasonably uniform deposit across the filter surface. The differences between duplicates with the acid treatment ranged from 5.9% to 11.8% for TC, from 4.5% to 13.4% for OC, and from 2.9% to 11% for EC. Differences between replicates are influenced by rinsing losses due to the formation of colloidal suspension and the small uneven distribution of sample residues on the filter. Differences due to the thermal/optical analysis are expected to be low because, at the end of each analysis, a 5% CH_4/He standard was injected for calibration (Han et al., 2007). An inter-laboratory comparison of IMPROVE with the DRI Model 2001 instrument with the (thermal manganese dioxide oxidation) TMO methods (by AtmAA, Inc., Calabasas, CA, USA) has shown good results (the difference was <5% for TC and 10% for OC and EC). Twelve blank filters were also analyzed, and the sample results were corrected by the average of the blank concentrations, which were 3.03, 2.99, and 0.04 μg m^{-3} for TC, OC, and EC, respectively. Particular quality assurance/quality control (QA/QC) procedures were described in Cao et al. (2003).

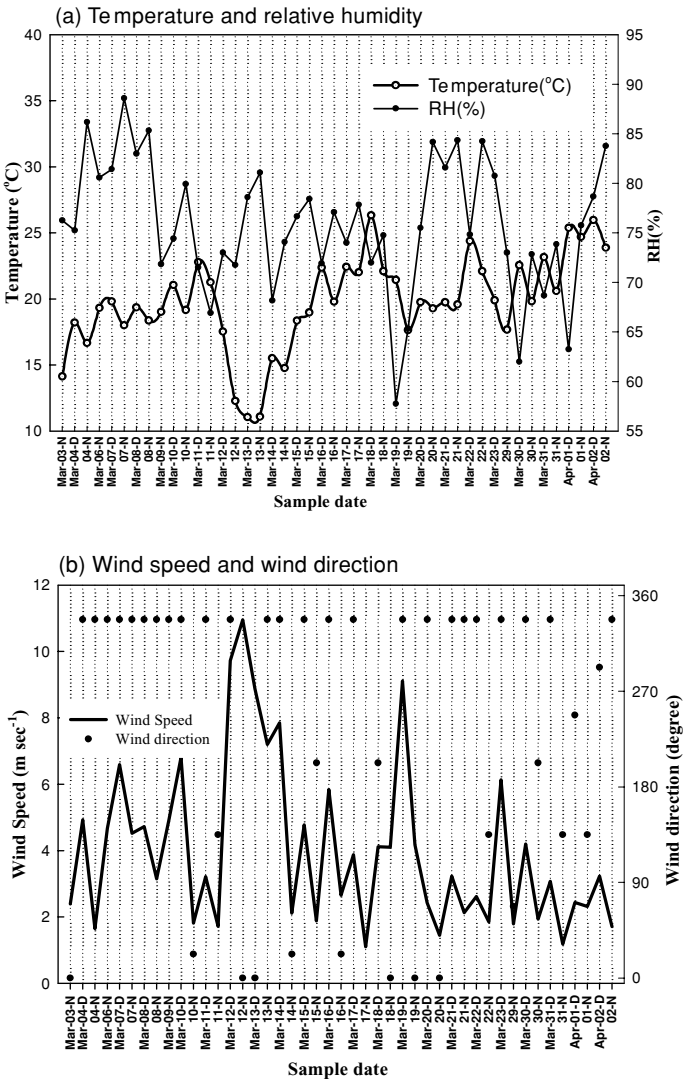


Figure 2. The time series of meteorological parameters during the sampling period. D, day samples; N, night samples.

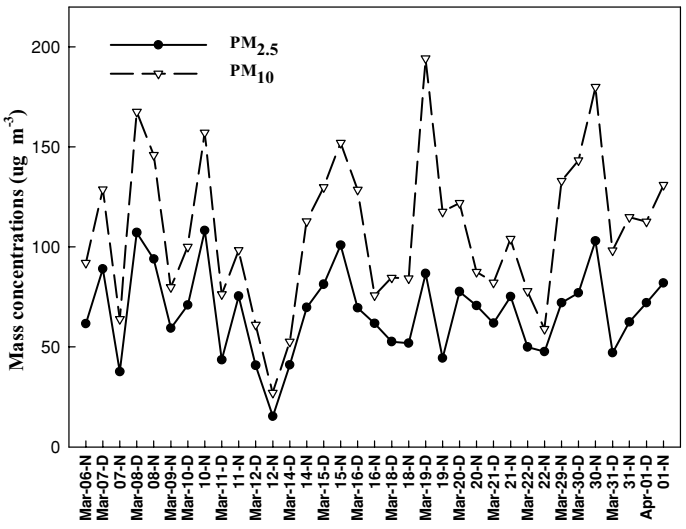


Figure 3. Average mass concentrations for fine ($PM_{2.5}$) and inhalable (PM_{10}) particulates at the Hungkuang University (HK) sampling site.

Results and Discussion

Meteorological Conditions and Ambient Air Particle Information

The sampling information (average temperature and relative humidity, wind speed, prevailing wind) about ambient air particulate concentrations is displayed in Figure 2. The average values for temperature, relative humidity, and wind speed are 20.1 °C, 74.8 %, and 4.0 m sec^{-1}, respectively at this near–highway-traffic sampling site during March 6 to April 1, 2006. The prevailing wind was blowing directly from the north-northwest wind during the sampling period in this study.

Mass Concentrations of Particulate Average Concentrations

Figure 3 shows the mass concentrations of $PM_{2.5}$ and PM_{10} particulate average concentrations at the HK sampling site during March to April 2006. The mass average concentrations of $PM_{2.5}$ and PM_{10} particulate were 67.5 μg $m^{-3}$and 107.9 μg m^{-3}, respectively, for during sampling period. Moreover, the $PM_{2.5}$ concentrations had lower concentration (15.2 μg m^{-3}) on March 12. Also, the PM_{10}particle concentrations showed highest concentrations (194.3 μg m^{-3}) on March 19, and lowest concentration (27.0 μg m^{-3}) occurred on March 12 during this sampling period. The result showed the concentrations of $PM_{2.5}$ and PM_{10} particles expressed the same distribution trend. A previous study has indicated that the average coarse ($PM_{2.5}$to PM_{10}) particle concentrations were approximately 19.5 mg m^{-3}, and the average $PM_{2.5}$ particle concentrations were 41.4 mg m^{-3}. The data obtained at the traffic road in previous research (Chang, et al., 2001) was approximately 0.7 km distance from HK sampling site in this study. In addition, the average $PM_{2.5}$ to PM_{10} particle concentrations in this study were approximately 2.1 times compared the previous study concentrations. Finally, the $PM_{2.5}$ particle concentrations obtained in this study were approximately 1.6 times that of the $PM_{2.5}$ particle concentration obtained in previous study (Chang et al., 2001).

Characterizations for Organic Carbon and Elemental Carbon Concentrations Variations

Figure 4 showed the average OC and EC concentrations of $PM_{2.5}$and PM_{10} particulate average concentrations at the HK sampling site. The average OC and EC concentrations for $PM_{2.5}$particulates were 10.22 and 3.88 μg m^{-3}, respectively, during sampling period. And the average OC and EC concentrations for PM_{10}particulates were 11.57 and 4.56 μg m^{-3}, respectively, during sampling period. Furthermore, the average EC concentrations obtained in the study for $PM_{2.5}$ particulates were 3.88 μg m^{-3}. This value was approximately 2.3 and 3.2 times compared with data obtained by traffic (Amsterdam, Netherlands) and on the outskirts (Ghent, Belgium), respectively (Viana et al., 2007).

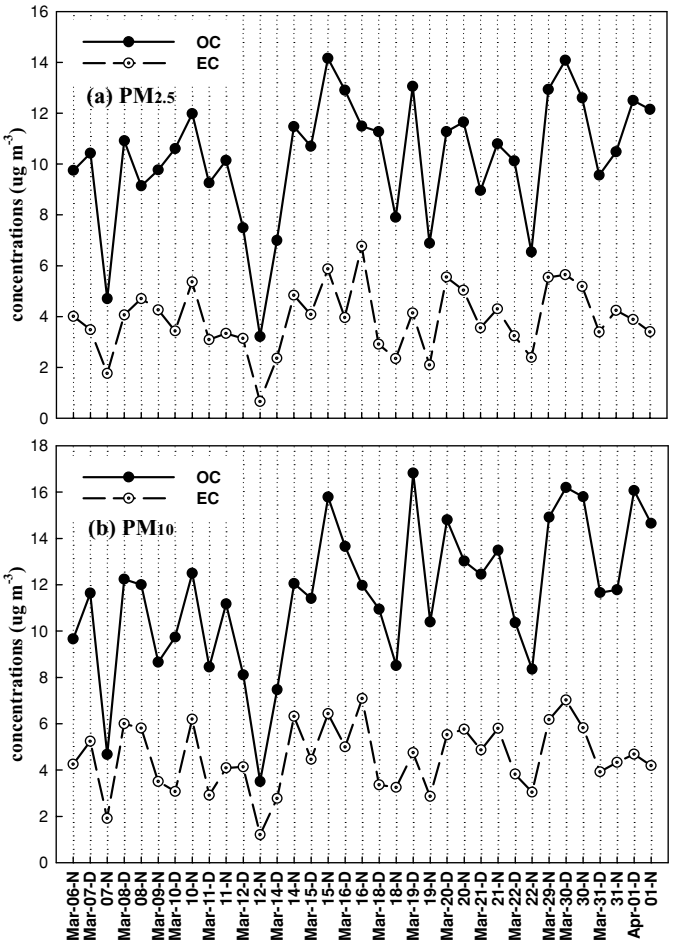


Figure 4. Sampling information of fine ($PM_{2.5}$) and inhalable (PM_{10}) particulates for EC and OC at the Hungkuang University (HK) sampling site.

Furthermore, the average ratios of OC/EC in $PM_{2.5}$ and PM_{10} particle concentrations were approximately 2.6 during sampling period at HK sampling sites. EC, which has a chemical structure similar to impure graphite, originates primarily from direct emissions of particles, predominantly during combustion. OC, from primary anthropogenic sources and from formation by chemical reactions in the atmosphere, rendered the concentrations of OC higher than EC at sampling site. The majority of carbonaceous aerosol was in the PM_{10} fraction. The average OC to EC concentrations ratios of $PM_{2.5}$ and PM_{10} are all larger than 1. These analytical results also reflected that OC concentration of mass concentrations was the major species at HK.

The OC and EC concentration expressed similar concentration pattern in central Taiwan for traffic and harbor sampling sites in this study. The result indicated similar average concentrations of large particulate and $PM_{2.5}$ particulate for expressed the same distribution trend, during sampling season. In addition, the average ratios of $PM_{2.5}$ to PM_{10} particles for mass concentrations were 1.83 at the TH sampling site. And the average mass concentrations ratios of $PM_{2.5}$ to PM_{10} are all larger than 1. The result displayed that the $PM_{2.5}$particulate concentration out of total mass concentrations was the major species at the TH sampling site. Finally, the average OC and EC concentrations in $PM_{2.5-10}$ and $PM_{2.5}$ for the TH were 28.8 and 4.19 μg m^{-3}, 6.50 and 1.06 μg m^{-3}, respectively, for the sampling period. The majority of carbonaceous aerosol was in the $PM_{2.5-10}$ fraction. In addition, the results also indicated that the average ratios of OC to EC for $PM_{2.5}$ concentrations were 8.72 at the TH sampling site. The average OC to EC concentrations ratios of $PM_{2.5}$ to PM_{10} are all larger than 1. The results also reflected that OC concentration out of mass total concentrations was the major species at the TH sampling site. The average OC concentrations at HK sampling site were approximately 1.6 times as that of TH sampling site. The average EC concentrations at HK sampling site were approximately 3.0 times as that of TH sampling site. The average concentrations ratios of OC/EC in $PM_{2.5}$ particle concentrations were approximately 2.6 during sampling period at HK sampling sites. The average concentrations ratios of OC/EC in $PM_{2.5}$ particle concentrations were approximately 4.9 during sampling period at TH sampling sites. T statistic was employed in this study at HK and TH sampling sites. The critical value α is equal $t_\alpha = 1–0.95 = 0.05$, sampling number $n = 17$, and t-test value is between ($-2.037 \leqq t_{17} \leqq 2.037$). Statistical analysis of average $PM_{2.5}$ concentration for the OC yielded a T statistic of 4.606, suggesting that the sample population means were significant differences. As for EC concentration in $PM_{2.5}$, the result indicated that the yielded a T statistic of 6.954, which is $> t_{\alpha/2,16} = 2.037$, suggesting that the sample population means were significant differences for both sampling periods.

The results indicated that the ratios of OC/EC ranged from 2.0 to 3.9 for $PM_{2.5}$ particles during the daytime sampling period at HK sampling site. The ratios of OC/EC in PM_{10} particles ranged from 2.0 to 3.5 during the daytime sampling period. The analytical results indicated ratios of OC/EC for $PM_{2.5}$particles ranged from 1.7 to 4.9 during the nighttime sampling period. The OC/EC ratios values ranged from 1.7 to 3.5 for PM_{10} particles during the nighttime sampling period. In general, the average ratios of OC/EC were higher (average ratios approximately 3.5) in the daytime than that of nighttime sampling periods for either PM_{10} or $PM_{2.5}$ particles.

Figure 5 displayed the OC/EC ratio of both PM_{10} and $PM_{2.5}$ particulates were at all times higher than 1.7, indicating that OC constituted 66–79% of TC. The daytime OC/EC ratio was higher than at nighttime, demonstrating that daytime direct emissions of primary organic aerosol (POA) are greater than emissions of carbonaceous material (EC aerosol) for either PM_{10} or $PM_{2.5}$ particles. In contrast, the nighttime OC/EC ratio was higher than at daytime, which has heavy traffic loadings, SOA was formed in the atmosphere through chemical reactions from reactive organic gases and subsequent gas-to-particle partitioning processes contributed to the nighttime aerosol in addition to the contribution from primary organic aerosol (POA) (Tsai and Chen, 2006; Turpin et al., 1991, 2000).

Estimate of the Secondary Organic Carbon

Table 1 showed the composition analysis of OC and EC out of total carbon for $PM_{2.5}$ and PM_{10} particulates at HK and TH

Table 1. *Composition analysis of organic carbon (OC) and elemental carbon (EC) of total carbon (TC) for fine ($PM_{2.5}$) and inhalable (PM_{10}) particulates at Hungkuang University (HK) and Taichung Harbor (TH) sampling site, respectively*

Sample category	Sample site	Mass ($\mu g\ m^{-3}$)	OCsec/Mass (%)	OCsec/OC (%)	OCsec/TC (%)	OCpri/TC (%)	EC/TC (%)
$PM_{2.5}$	HK	67.4	5.9	36.3	27.0	46.0	27.0
	TH	39.9	13.3	79.9	70.0	15.7	14.3
PM_{10}	HK	107.8	3.7	32.8	24.1	47.8	28.1
	TH	62.2	43.8	77.2	70.3	17.4	12.3

OCpri, primary organic carbon; OCsec, secondary organic carbon.

sampling site, respectively. This estimation was calculated by using the method suggested by Turpin and Huntzicker (1995).

Turpin and Huntzicker (1995) suggested the following equation to identify the secondary organic carbon (SOC) based on the hypothesis noted previously:

$$\text{OCsec} = \text{OCtol} - \text{EC} \times (\text{OC/EC})\text{pri} \quad (1)$$

where OCsec is the SOC, and OCtol the measured ambient total OC. The ratio (OC/EC)pri varies with various sources and the assumed prime ratio. Castro et al. (1999) suggested that (OC/EC)pri could be replaced by (OC/EC)min if the (OC/EC)min contain exclusively the primary carbonaceous compounds, and the former equation could be written as follows:

$$\text{OCsec} = \text{OCtol} - \text{EC} \times \text{OC/EC})\text{min}. \quad (2)$$

Over the study period, the average concentrations of SOC in $PM_{2.5}$ and PM_{10} for the HK sampling site ranged from 1.8 to 6.4 $\mu g\ m^{-3}$ with a mean value of 3.6 $\mu g\ m^{-3}$ and from 1.1 to 8.7 $\mu g\ m^{-3}$ with a mean value of 3.8 $\mu g\ m^{-3}$, respectively. In addition, the average SOC in $PM_{2.5}$ and PM_{10} were concentrations ranging from 2.6 to 15.3 $\mu g\ m^{-3}$, with means of 5.3 $\mu g\ m^{-3}$, from 8.9 to 49.2 $\mu g\ m^{-3}$ and 27.8 $\mu g\ m^{-3}$, respectively, at the TH sampling site. The results indicated that the average composition ratios in $PM_{2.5}$ and PM_{10} of primary OC out of total carbon were 46.0% and 47.8%, respectively at HK. And the results indicated that the average composition ratios in $PM_{2.5}$ and PM_{10} of secondary primary OC out of total carbon were 27.0 and 24.1%, respectively, at HK.

As shown in Figure 6, correlation coefficients for OC/EC were both greater than 0.6 in either $PM_{2.5}$ or PM_{10} at this

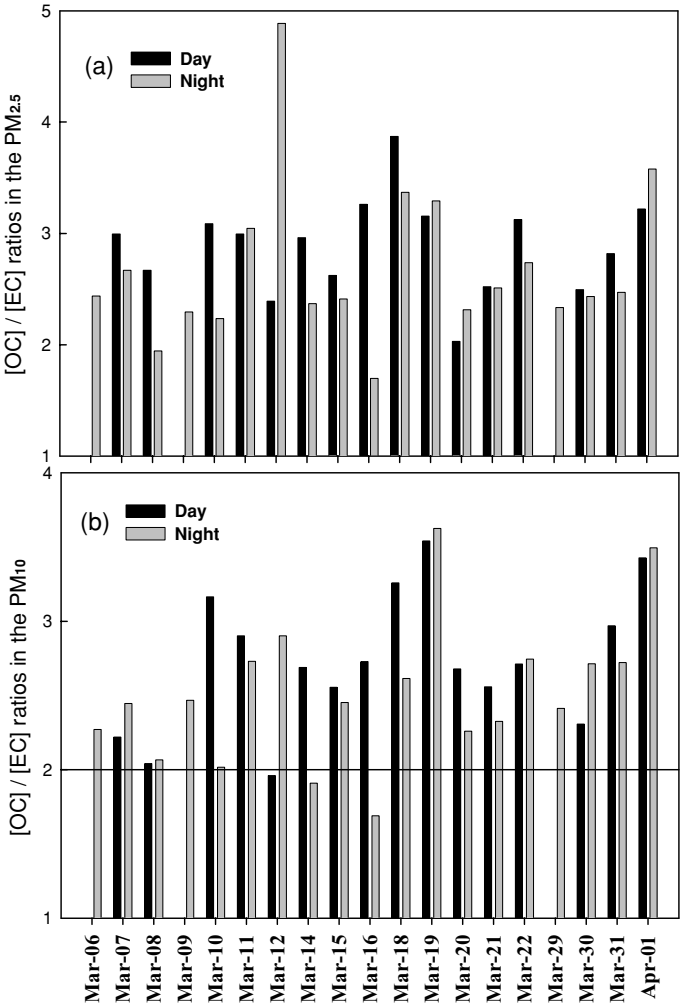


Figure 5. Ratio of organic carbon (OC)/elemental carbon (EC) in a) fine ($PM_{2.5}$) particulates and b) inhalable (PM_{10}) particulates during the daytime and nighttime sampling periods.

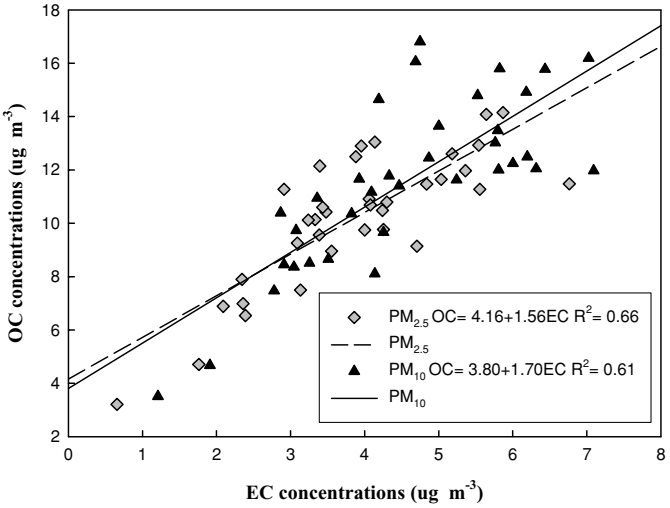
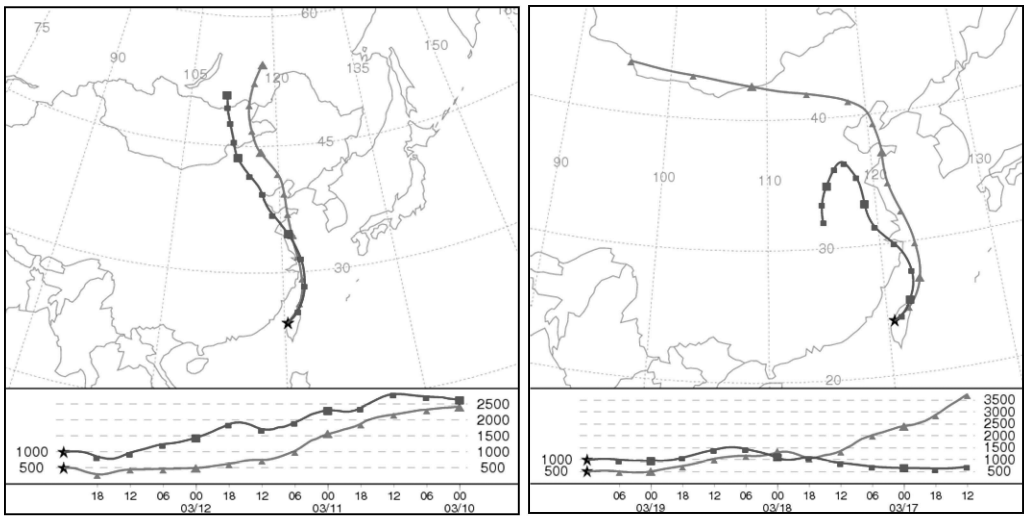


Figure 6. The relationship between organic carbon (OC) and elemental carbon (EC) concentrations in fine ($PM_{2.5}$) and coarse (PM_{10}) particulates at the Hungkuang University (HK) sampling site.



(a) Nighttime sampling period, 12 March 2006 (b) Daytime sampling period, 19 March 2006

Figure 7. Result of the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model with 3-day backward trajectory analysis at heights of 500, and 1000 m at the Hungkuang University (HK).

near–highway-traffic sampling site. The regression result (solid) for $PM_{2.5}$ particulates displayed a slope and intercept of 4.2 and 1.6, respectively. In addition, the regression result (solid) for PM_{10} particulates displayed a slope and intercept of 3.8 and 1.7, respectively. The fitting results of a single linear regression revealed a common source, which explains only 60% of the variance. Alternatively, we can explain the variance as the mixture of two primary sources, with characteristic ratios given by the edges of the data spread, as shown by the two dashed lines in Figure 6 (Husain et al., 2007).

Air Mass Trajectories

Three-day back trajectories were calculated by Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler, 1999) with 6-hourly archived meteorological data provided from the United States (US) National Centers for Environmental Prediction (NCEP) global data assimilation system (GDAS) was applied in this study, developed by the US National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory. The location of backward trajectory was started at Shalu with the altitude of 500 m and 1000 m AGL. The trajectory computations were carried out two times daily from March 6 to April 1, 2006, with the start time of 00 and 12 coordinated universal time (UTC). Run time of every trajectory was used to obtain 72 h (3 days) backward air mass trajectories. FNL meteorological datasets were used to compute trajectories, which included the sampling site and arrival time of input.

Experimental results indicated that high wind speed event was occurred on 0312N. This kind of event was originated from Mainland China of the dust storm. Its trajectories can be extended to the yellow sand area of Mainland China. Its trajectories process occurred at the height of mixed layer. The trajectories path occurred below the mixed layer once it entered the ocean area. Thus, the impact of the ambient air pollutants to Taiwan is insignificant (Figure 7a).

In another regard, analytical results also indicated that another high-wind speed event occurred on 0319D. This kind of event originated from Mainland China of the dust storm. Its trajectories can be extended to the yellow sand area of Mainland China. However, its trajectories' process occurred below the height of mixed layer. Thus, this process can bring the air pollutants from Mainland China directly to Taiwan (Figure 7b).

Conclusions

The observed results displayed the concentrations of $PM_{2.5}$ and PM_{10} particles, which expressed the same distribution trend during sampling period. In addition, the average mass concentrations ratio of $PM_{2.5}$ to PM_{10} particulates was 1.6 at the HK sampling site. And the average particulates concentrations ratios of $PM_{2.5}$ to PM_{10} particulate are all larger than 1 during the whole sampling period. Experimental results also reflected that $PM_{2.5}$particulate concentrations were the major species (average roughly 61%) at the HK sampling site. Furthermore, the results reflected that the primary OCs mainly existed by approximately 47% at HK (traffic area) sampling site for either $PM_{2.5}$ or PM_{10} particulates. However, the secondary OCs were mainly approximately 70% for either $PM_{2.5}$ or PM_{10} particulates at TH (harbor area).

Finally, the average OC concentrations in PM_{10} and $PM_{2.5}$ for HK were 11.68 and 10.01 $\mu g\ m^{-3}$ during the daytime sampling period, respectively. And the average EC concentrations of PM_{10} and $PM_{2.5}$ at this sampling site were 4.23 and 3.57 $\mu g\ m^{-3}$, respectively, during the daytime sampling period. Moreover, the average OC concentrations in PM_{10} and $PM_{2.5}$ during the nighttime were 11.18 and 9.56 $\mu g\ m^{-3}$, respectively, at HK. Furthermore, the average EC concentrations of PM_{10} and

$PM_{2.5}$ at this sampling site were 4.62 and 3.88 $\mu g\ m^{-3}$, respectively, during the nighttime sampling period. Analytical results also demonstrated that the majority of the carbonaceous aerosol was in the $PM_{2.5}$ particulate fraction. Additionally, this result also reflected that OC was the major species for all particulate sizes.

Acknowledgement

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