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Effects of day-of-week trends and vehicle types on $PM_{2.5}$-bounded carbonaceous compositions

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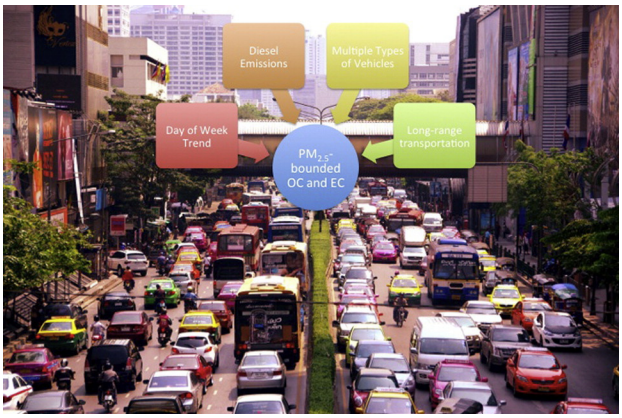
HIGHLIGHTS

- Traffic emissions play an important role in governing OC and EC during weekdays.
- Time series analysis shows the existence of day-of-week trends of OC and EC.
- Diesel vehicles are the main contributors of carbonaceous compositions.

GRAPHICAL ABSTRACT



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ABSTRACT

Carbonaceous compositions of $PM_{2.5}$ were measured in the heart of Bangkok from 17th November 2010 to 19th January 2012, and a data set of 94 samples was constructed. Effects of day-of-week trends and vehicle types on $PM_{2.5}$-bound TC, OC, and EC were carefully investigated. In this study, OC was the most important contributor to the total $PM_{2.5}$ mass concentration. The average $PM_{2.5}$-bound OC content measured at CHAOS ($18.8 \pm 9.18\ \mu g\ m^{-3}$) was approximately 11 times higher than at Chaumont, Switzerland ($1.7\ \mu g\ m^{-3}$), but approximately five times lower than at Xi'an, China ($93.0\ \mu g\ m^{-3}$). The application of diagnostic binary ratios of OC/EC and estimations of secondary organic carbon (SOC) coupled with autocorrelation plots (Box and Jenkins) highlight the enhanced impacts of traffic emissions, especially from diesel vehicles, on $PM_{2.5}$-bound carbonaceous compositions on weekdays relative to weekends. Hierarchical cluster analysis (HCA) coupled with principal component analysis (PCA) underline the importance of diesel emissions as the primary contributors of carbonaceous aerosols, particularly during weekdays.

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1. Introduction

Chemical characteristics of aerosols, particularly the compositions of organic carbon (OC) and elemental carbon (EC), have been constantly evaluated predominantly in Asian countries during the past few years (Pongpiachan et al., 2013a,2013b, 2014a,2014b; Srivastava et al., 2014; Zhang et al., 2009). Rapid industrialization and urbanization, especially in China, are apparently responsible for the enhancement of carbonaceous aerosols in this region (Huang et al., 2013; Zhang et al., 2011). Recently, numerous studies have highlighted the impacts of burning agricultural waste and forest fires in Southeast Asian countries and southern China as the main contributors of OC and EC in fine particles (Pongpiachan et al., 2014a; Zhang et al., 2010). Previous studies have shown associations between carbonaceous aerosols and the number of patients with respiratory diseases as well as some major contributions on the climate system (Künzi et al., 2013; Repine et al., 2008; US-EPA, 2012).

As a consequence, many scientific studies have focused on annual average, monthly and seasonal trends in the concentrations of carbonaceous compositions (Fu et al., 2014; Tao et al., 2012), which are extremely advantageous to a comprehensive view on the average contributions of sources of particulate OC and EC. However, it is extremely difficult to investigate the influences of day-of-week trends only given knowledge of annual or monthly average carbonaceous concentrations. In most incidents, the impacts of numerous source emissions coupled with photochemical transformations can greatly affect the day-of-week trends of atmospheric pollutants (Chow, 2003). Furthermore, traffic emissions seem to govern the diurnal variations and weekly trends of OC and EC (Bae et al., 2004). While others have examined the effects of anthropogenic activities on both diurnal and weekly fluctuations, only a few studies have demonstrated in-depth, quantitative evidence associated with the behaviour of carbonaceous aerosols in tropical countries (Li et al., 2012; Pongpiachan et al., 2009; Safai et al., 2014).

Despite copious studies involving the clarification of emission factors of OC/EC compositions from different fuel and vehicle types (Shen et al., 2014; Wei et al., 2014), little is known about their correlations in the tropical atmosphere. Although vehicle conditions can play a major role in the emission source strength of carbonaceous aerosols, there have been no studies to date that examine the relationship between vehicle types and OC/EC compositions. To our knowledge, there is no information available on intensive monitoring of fine particle bounded carbonaceous compositions in Thailand. Overall, the principle objectives of this study were to (*i*) assess the day-of-week trends of OC/EC compositions through an intensive monitoring campaign of $PM_{2.5}$ and (*ii*) elucidate the correlations between vehicle types and carbonaceous aerosols in the heart of metropolitan Bangkok, Thailand.

2. Experiment

2.1. Description of sampling sites

In this study, the roof of the Mahamakut Building, Chulalongkorn University (CHAOS: 13°44′9.07″N 100°31′50.26″E), was selected as the air quality observatory site because the site is positioned in the most densely populated area of Bangkok and is surrounded by shopping malls, restaurants, multiplex movie theatres, and business buildings (see Fig. 1). There were no barriers in the neighbourhood of the sampling equipment, which was deliberately situated to be accessible to winds from all directions. The sampling method was based on the "Quality Assurance Guidance Document 2.12; Monitoring $PM_{2.5}$ in Ambient Air Using Designated Reference or Class I Equivalent Methods" (US-EPA, 1998).

2.2. Filter sample collection & meteorological data

To ensure that the quantity of carbonaceous compositions present in the samples was much greater than the instrumental detection limit, MiniVol™ portable air samplers (Airmetrics) were employed to collect $PM_{2.5}$ for 72 h at CHAOS. The MiniVol's pump draws air at 5 L min^{-1} through a particle size separator (impactor) and then through a 47 mm filter. The 2.5-micron particle ($PM_{2.5}$) separation is achieved by impaction. In this study, $PM_{2.5}$ samples were collected on 47 mm Whatman quartz microfiber filters (QM/A) for the chemical detection of OC/EC compositions. All samples ($n = 94$) were collected in three consecutive day intervals from 17th November 2010 to 19th January 2012 at CHAOS. Prior to the $PM_{2.5}$ sample collection, the QM/A were baked at 800 °C for at least three hours to remove any organic contaminants and were wrapped individually in DCM pre-cleaned aluminium foil until loaded into the filter holder cassette. The QM/A were equilibrated for 24 h at a constant temperature between 20 °C and 23 °C and *RH* between 35% and 45% in a laboratory at the Institute of Earth Environment, Chinese Academy of Sciences (IEECAS), Xi'an, China, prior to $PM_{2.5}$ mass measurement.

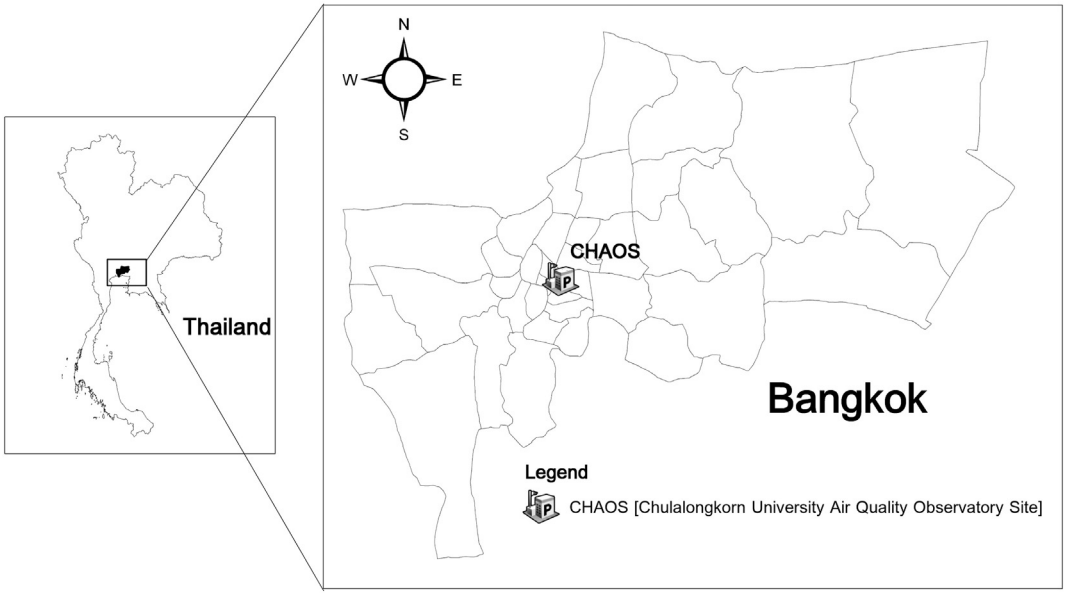

Fig. 1. Map of Chulalongkorn University Air Quality Observatory Site (CHAOS).

Table 1
Average OC and EC concentrations and the OC/EC ratios in $PM_{2.5}$ collected at CHAOS.

Sampling date	$PM_{2.5}$ [μg m^{-3}]	TC [μg m^{-3}]	OC [μg m^{-3}]	EC [μg m^{-3}]	OC/EC ratio	TC [%]	OC [%]	EC [%]
11/17/10	39.24	17.84	11.17	6.67	1.68	45.5	28.5	17.0
11/20/10	54.72	35.59	24.40	11.19	2.18	65.0	44.6	20.4
11/23/10	35.21	18.29	11.91	6.37	1.87	51.9	33.8	18.1
11/26/10	27.08	17.34	12.89	4.45	2.90	64.0	47.6	16.4
12/02/10	35.62	12.39	8.18	4.21	1.94	34.8	23.0	11.8
12/05/10	55.69	23.95	16.83	7.11	2.37	43.0	30.2	12.8
12/08/10	60.97	25.22	16.71	8.51	1.97	41.4	27.4	13.9
12/11/10	30.89	10.25	6.64	3.62	1.84	33.2	21.5	11.7
12/14/10	25.56	15.29	9.78	5.51	1.77	59.8	38.3	21.6
12/17/10	39.03	22.75	17.63	5.11	3.45	58.3	45.2	13.1
12/20/10	75.35	45.13	33.04	12.09	2.73	59.9	43.9	16.0
12/23/10	62.57	37.67	23.86	13.81	1.73	60.2	38.1	22.1
12/26/10	32.78	16.03	11.47	4.56	2.51	48.9	35.0	13.9
12/29/10	68.68	43.65	31.30	12.36	2.53	63.6	45.6	18.0
01/01/11	58.12	29.69	22.05	7.65	2.88	51.1	37.9	13.2
01/04/11	49.86	30.67	21.19	9.48	2.23	61.5	42.5	19.0
01/07/11	37.29	20.95	15.03	5.93	2.54	56.2	40.3	15.9
01/10/11	54.37	32.95	23.59	9.36	2.52	60.6	43.4	17.2
01/13/11	47.08	27.09	18.94	8.15	2.32	57.5	40.2	17.3
01/16/11	53.26	23.50	17.66	5.84	3.03	44.1	33.2	11.0
01/19/11	74.17	47.79	33.49	14.30	2.34	64.4	45.2	19.3
01/22/11	52.99	44.04	32.78	11.26	2.91	83.1	61.9	21.3
01/25/11	49.86	32.21	23.32	8.89	2.62	64.6	46.8	17.8
01/28/11	40.07	32.27	23.71	8.56	2.77	80.5	59.2	21.4
01/31/11	57.22	44.01	32.07	11.94	2.68	76.9	56.0	20.9
02/03/11	58.54	44.96	33.28	11.68	2.85	76.8	56.8	19.9
02/06/11	52.71	28.39	21.43	6.96	3.08	53.9	40.7	13.2
02/09/11	33.68	15.74	10.40	5.33	1.95	46.7	30.9	15.8
02/12/11	32.50	9.90	6.37	3.53	1.81	30.5	19.6	10.9
02/15/11	35.35	14.14	7.68	6.46	1.19	40.0	21.7	18.3
02/18/11	29.72	13.22	9.10	4.12	2.21	44.5	30.6	13.9
02/21/11	41.88	20.86	13.07	7.79	1.68	49.8	31.2	18.6
02/24/11	24.72	6.70	3.73	2.96	1.26	27.1	15.1	12.0
02/27/11	33.61	27.50	21.46	6.05	3.55	81.8	63.8	18.0
03/02/11	57.36	25.99	17.16	8.83	1.94	45.3	29.9	15.4
03/05/11	74.31	38.20	27.62	10.58	2.61	51.4	37.2	14.2
03/08/11	27.36	7.65	4.71	2.93	1.61	27.9	17.2	10.7
03/11/11	62.92	39.62	30.55	9.07	3.37	63.0	48.6	14.4
04/27/11	20.63	3.97	1.93	2.04	0.94	19.3	9.3	9.9
04/30/11	18.82	6.55	3.88	2.67	1.46	34.8	20.6	14.2
05/03/11	15.35	5.04	2.85	2.19	1.30	32.8	18.5	14.3
05/05/11	15.97	6.88	5.33	1.54	3.46	43.0	33.4	9.6
08/13/11	67.43	37.55	29.58	7.97	3.71	55.7	43.9	11.8
08/16/11	48.68	18.26	14.31	3.94	3.63	37.5	29.4	8.1
08/19/11	54.58	19.09	15.32	3.76	4.07	35.0	28.1	6.9
08/22/11	49.79	16.54	12.36	4.18	2.96	33.2	24.8	8.4
08/25/11	43.82	20.80	14.85	5.96	2.49	47.5	33.9	13.6
08/28/11	20.21	11.20	7.71	3.50	2.20	55.4	38.1	17.3
08/31/11	74.31	46.88	36.66	10.22	3.59	63.1	49.3	13.8
09/03/11	58.54	38.91	30.02	8.89	3.38	66.5	51.3	15.2
09/06/11	66.74	40.39	31.80	8.59	3.70	60.5	47.6	12.9
09/09/11	60.90	35.98	29.25	6.73	4.35	59.1	48.0	11.0
09/12/11	50.07	21.10	15.83	5.28	3.00	42.1	31.6	10.5
09/15/11	45.49	21.40	15.69	5.70	2.75	47.0	34.5	12.5
09/30/11	13.82	8.65	8.30	3.60	2.30	62.6	60.0	2.6
09/24/11	41.94	25.37	16.95	8.42	2.01	60.5	40.4	20.1
09/27/11	33.82	21.66	17.43	4.24	4.11	64.1	51.5	12.5
09/18/11	53.26	28.78	21.34	7.44	2.87	54.0	40.1	14.0
10/03/11	69.65	40.16	29.90	10.25	2.92	57.7	42.9	14.7
10/06/11	74.24	45.67	33.99	11.68	2.91	61.5	45.8	15.7
10/09/11	87.78	57.46	43.00	14.46	2.97	65.5	49.0	16.5
10/12/11	79.24	51.24	39.86	11.38	3.50	64.7	50.3	14.4
10/15/11	53.47	25.90	19.74	6.16	3.20	48.4	36.9	11.5
10/18/11	46.18	22.21	17.46	4.76	3.67	48.1	37.8	10.3
10/21/11	34.10	13.54	9.90	3.65	2.72	39.7	29.0	10.7
10/24/11	28.54	26.26	19.77	6.49	3.05	92.0	69.3	22.7
10/27/11	48.33	18.29	13.10	5.19	2.53	37.8	27.1	10.7
10/30/11	68.26	38.94	31.03	7.91	3.92	57.0	45.5	11.6
11/02/11	62.43	33.16	25.49	7.68	3.32	53.1	40.8	12.3
11/05/11	35.90	28.27	22.23	6.05	3.68	78.7	61.9	16.8
11/08/11	53.96	24.18	18.20	5.99	3.04	44.8	33.7	11.1
11/11/11	52.15	27.26	20.74	6.52	3.18	52.3	39.8	12.5
11/14/11	54.24	23.35	17.28	6.08	2.84	43.1	31.9	11.2
11/17/11	45.14	21.01	15.83	5.19	3.05	46.5	35.1	11.5
11/20/11	55.42	24.42	18.08	6.34	2.85	44.1	32.6	11.4
11/23/11	64.44	35.56	26.20	9.36	2.80	55.2	40.7	14.5

Table 1 (*continued*)

Sampling date	$PM_{2.5}$ [μg m^{-3}]	TC [μg m^{-3}]	OC [μg m^{-3}]	EC [μg m^{-3}]	OC/EC ratio	TC [%]	OC [%]	EC [%]
11/26/11	38.33	17.60	12.54	5.07	2.47	45.9	32.7	13.2
11/29/11	58.96	35.41	28.92	6.49	4.46	60.1	49.1	11.0
12/02/11	36.67	18.17	14.20	3.97	3.57	49.5	38.7	10.8
12/05/11	46.18	14.64	11.20	3.44	3.26	31.7	24.3	7.4
12/08/11	41.67	12.39	9.10	3.29	2.77	29.7	21.8	7.9
12/11/11	42.36	14.46	10.16	4.30	2.37	34.1	24.0	10.1
12/14/11	42.01	27.77	22.43	5.33	4.21	66.1	53.4	12.7
12/17/11	27.92	14.28	11.17	3.11	3.59	51.2	40.0	11.1
12/20/11	54.03	27.81	21.63	6.18	3.50	51.5	40.0	11.4
12/23/11	61.04	29.69	23.41	6.28	3.73	48.6	38.4	10.3
12/26/11	59.79	30.51	24.40	6.10	4.00	51.0	40.8	10.2
12/29/11	41.60	16.92	13.31	3.62	3.68	40.7	32.0	8.7
01/04/12	59.44	25.46	18.64	6.82	2.73	42.8	31.4	11.5
01/07/12	9.24	6.12	5.91	2.10	2.82	66.3	64.0	2.2
01/10/12	39.58	16.71	11.71	5.01	2.34	42.2	29.6	12.7
01/13/12	39.65	17.84	13.31	4.53	2.93	45.0	33.6	11.4
01/16/12	59.44	30.44	23.92	6.52	3.67	51.2	40.2	11.0
01/19/12	70.42	37.40	29.43	7.97	3.69	53.1	41.8	11.3
Average	47.6 ± 16.4	25.4 ± 11.9	18.8 ± 9.18	6.65 ± 2.94	2.80 ± 0.77	52.1 ± 13.6	38.4 ± 11.8	13.6 ± 4.00

2.3. Organic carbon (OC) & elemental carbon (EC)

A 0.5-cm^2 punch from each QM/A was examined for carbonaceous compositions with a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) for four organic carbon (OC) fractions (OC1, OC2, OC3, and OC4) in a helium atmosphere, three elemental carbon (EC) fractions (EC1, EC2, and EC3) in a 2% oxygen–98% helium atmosphere, and OP, a pyrolysed carbon fraction following the IMPROVE (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Chow et al., 2001; Fung et al., 2002). For this study, OC and EC were described as the sum of OC fractions (OC1 + OC2 + OC3 + OC4) and EC fractions (EC1 + EC2 + EC3 + OP), respectively, based on the IMPROVE TOC (Interagency Monitoring to Protect Visual Environments Total Organic Carbon) protocol (Chow et al., 2001; Fung et al., 2002). Total organic carbon (TOC) was defined as the sum of OC and EC. The quality control and quality assurance (QA/QC) protocols have been previously discussed in detail in Cao et al. (2003).

2.4. Traffic count database and statistical analysis

The Traffic and Transportation Department (TTD), Bangkok Metropolitan Administration (BMA), provided all traffic data used in this study. To formulate effective policy for traffic volume control and develop the traffic and transportation information system for planning and public dissemination, TTD strategically determined 153 traffic flow observatory sites for counting the number of vehicles based on their corresponding category, namely pickups/vans, trucks, cars, buses and Tuktuks (i.e., a three wheel motorcycle). The data synchronization between the two databases (i.e., vehicle counts and OC/EC compositions) was conducted manually for further advanced statistical analysis. In addition, the Statistical Program for Social Sciences (SPSS) version 13 was used for Simple Linear Regression Analysis (SLRA), Analysis of Variance (ANOVA), Hierarchical Cluster Analysis (HCA), and Principal Component Analysis (PCA).

2.5. Estimation of secondary organic carbon (SOC)

Because several factors can cause relatively high OC/EC ratios in ambient aerosols, it is crucial to conduct further estimation of secondary organic carbon (SOC), which is frequently associated with long-range transport (Wang et al., 2012; Zhou et al., 2012). In this study, the calculation of SOC was performed using the method reported by Na et al. (2004). The principle of this technique is based on the assumption that $PM_{2.5}$ samples that possess the smallest OC/EC ratios comprise almost entirely primary carbonaceous compositions (Castro et al., 1999). For the $PM_{2.5}$ samples detected at CHAOS, the average of the three lowest OC/EC ratios was 1.13 ± 0.17 and thus can be used to estimate SOC. It is also essential to note that the three lowest OC/EC ratios are assumed to have solely primary OC, and the influence of a small proportion of SOC is of minor importance. The concentration of SOC is calculated by

$$OC_{sec} = OC_{tot} - EC \times (OC/EC)_{primary}, \quad (1)$$

where OC_{sec}, OC_{tot}, and $(OC/EC)_{primary}$ are SOC, TOC, and the average value of the three lowest OC/EC ratios, respectively.

2.6. Probability distribution function of carbonaceous compositions

The probability distribution function (PDF) was applied to $PM_{2.5}$-bound TC, OC, and EC collected at CHAOS. Generally, a PDF is a function that describes the relative probability for a random parameter to take a provided value. The Gaussian distribution describes the probability that a random parameter will fall within a specific interval

$$y = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(\frac{-(x-\mu)^2}{2\sigma^2}\right), \quad (2)$$

where y, σ, σ^2, μ and x are the probability distribution function, standard deviation, variance, average and concentrations of carbonaceous compositions, respectively.

2.7. Time series approach

During the past few years, autocorrelation plots have been widely employed in several atmospheric environmental studies, including an investigation of the impact of meteorological parameters and trace gas concentrations on daily hospital walk-ins and admissions in Chiang-Mai, Thailand (Pongpiachan and Paowa, 2014), long-term observations and modelling of aerosol loading over the Indo-Gangetic plains (IGP), India (Soni et al., 2014), and pedestrian exposure to $PM_{2.5}$ in Sydney, Australia (Greaves et al., 2008). Because autocorrelation represents the similarity between observations as a function of the time lag between them, it appears reasonable to evaluate the randomness of carbonaceous compositions with time using this mathematical tool. This randomness can be investigated by computing autocorrelations for each parameter at different time lags. In the case of random signals, such autocorrelations should hypothetically approach zero for all

time-lag intervals. In the case of non-random, one or more of the autocorrelations will be significantly non-zero. The calculation of autocorrelation plots is described below. Primarily, the vertical axis symbolizes the autocorrelation coefficient, which is computed from Eq. (3):

$$R_h = \frac{C_h}{C_0} \tag{3}$$

where R_h is the autocorrelation coefficient of the atmospheric parameters (i.e., $PM_{2.5}$, TC, OC, and EC) and ranges between −1 and +1. Note that C_h is the autocovariance function, which can be explained in Eq. (4):

$$C_h = \frac{1}{N}\sum_{t=1}^{N-h}(Y_t - \overline{Y})(Y_{t+h} - \overline{Y}) \tag{4}$$

where N, t, h, Y_t, $\overline{Y}$, Y_{t+h} are the total number of individual atmospheric parameters, time, time lag, concentration of the atmospheric parameter at time t, average of the atmospheric parameter concentrations, and concentration of the atmospheric parameter at time $t + h$, respectively. It is also important to note that C_0 is the variance function, which can be described as follows:

$$C_0 = \frac{\sum_{t=1}^{N}(Y_t - \overline{Y})^2}{N}. \tag{5}$$

Furthermore, the horizontal axis denotes the time lag h ($h = 1, 2, 3, \ldots$). Finally, the confidence bands have fixed width that depend on the sample size and can be computed using Eq. (6):

$$\pm\frac{Z_{1-\alpha/2}}{\sqrt{N}} \tag{6}$$

where N is the sample size, Z is the cumulative distribution function of the standard normal distribution, and α is the significance level.

3. Results & discussion

The statistical descriptions and their percentage contributions of $PM_{2.5}$, TC, OC and EC measured during the sampling period at CHAOS are listed in Table 1. The percentage contribution of TC ranged from 19.3 to 92%, with an average of 52.1 ± 13.6%, and OC varied from 9.3 to 69.3%, with an average of 38.4 ± 11.8%. EC ranged from 2.2 to 22.7%, with an average of 13.6 ± 4.0%. These findings show that OC was the most important contributor to the total $PM_{2.5}$ mass concentration.

3.1. OC/EC ratios and estimation of secondary organic carbon (SOC)

During the past decades, OC/EC ratios have been extensively applied for elucidating the photolysis process of carbonaceous compositions and the formation of secondary organic aerosol (SOA) and for categorizing their emission sources (Turpin and Huntzicker, 1995). In this study, the OC/EC ratios varied from 0.94 to 4.46, with an average of 2.80 ± 0.77, as listed in Table 1. Generally, the average OC/EC ratio at CHAOS was very similar to that of Chaumont, Switzerland (2.8), Guangzhou, China (2.8 ± 2.8), and Xi'an, China (2.9 ± 2.7) (Cao et al., 2003, 2005; Hueglin et al., 2005). Several reasons can be used to explain the comparatively high OC/EC ratios observed in this study. Firstly, previous studies have highlighted the importance of the formation of secondary OC via long-range transport (Wang et al., 2012; Zhou et al., 2012). For instance, the moderately high $PM_{2.5}$-bound OC/EC ratios (range: 1.6–10.4; average: 5.2 ± 1.8) observed at Mount Heng, China were considered to be a consequence of in-cloud secondary organic aerosol (SOA) formation coupled with long-range transport (Zhou et al., 2012). The increase in the photochemical age of the air mass during the spring and summer also played an important role in relatively high OC/EC ratios in the North China Plain (Wang et al., 2012). Alternatively, the fairly high OC/EC ratios simply can be attributed to the extraordinarily low EC

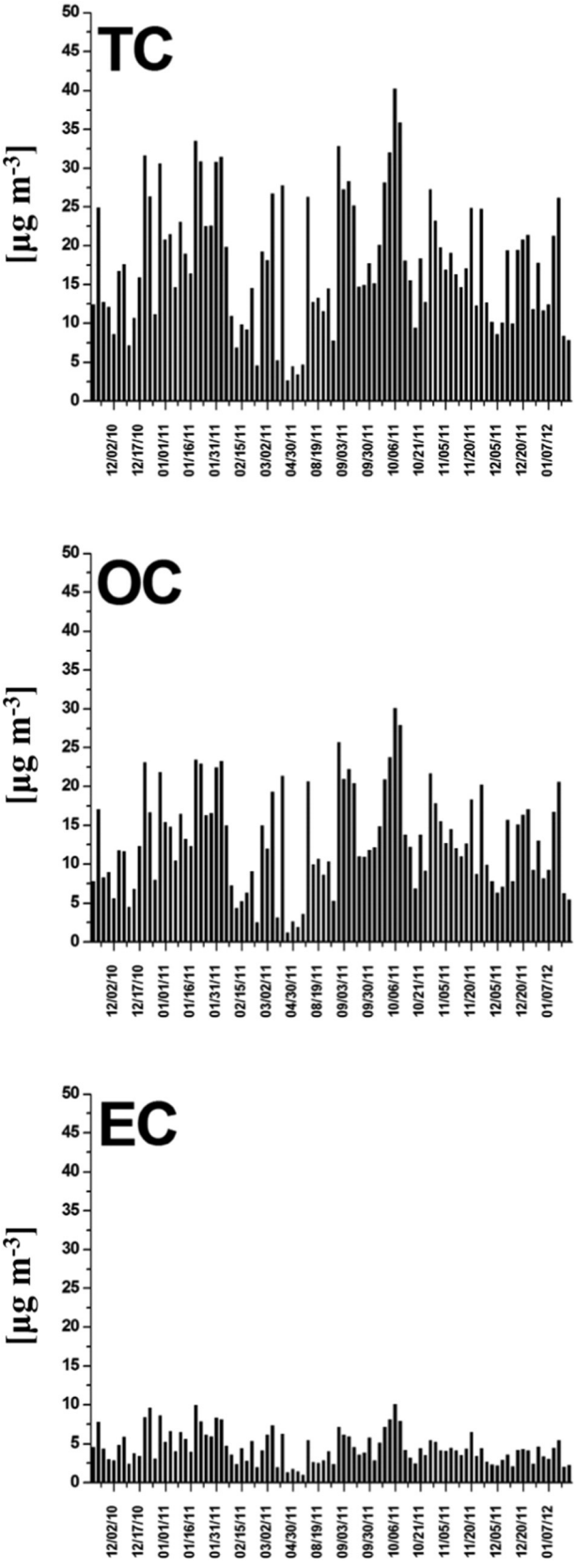

Fig. 2. Temporal variability in fine carbonaceous aerosol from November 2010 to January 2012 in CHAOS.

levels in rural areas, and thus the OC/EC ratio tended to be high. It is also important to note the probable relationship between biomass burning and the formation of SOA (Zeng and Wang, 2011). As a consequence, the burning of agricultural waste coupled with forest fires could have been responsible for the high OC/EC ratios.

Using Eq. (1), the $PM_{2.5}$-bound OC_{sec} concentrations and percentage contributions at CHAOS were 12.6 ± 6.20 μg m^{-3} and 68.9 ± 7.22%, respectively. The average percentage contribution was almost equivalent to that of Claremont, US $PM_{2.5}$ (65%, Na et al., 2004) but almost four times and 1.7 times higher than those of Birmingham, UK (17%, Castro et al., 1999) and Kaohsiung (40.0%, Lin and Tai, 2001), respectively. This emphasizes the importance of long-range transport as the primary mechanism responsible for the relatively high observed OC/EC ratios at CHAOS, and thus other mechanisms, such as local agricultural waste and biomass burning, can be considered negligible.

3.2. Day-of-week trends of OC/EC ratios and OC_{soc}

Over the past few years, the application of OC/EC ratios has been widely used for elucidating the relationship between day-of-week trends and vehicular emission sources (Chinkin et al., 2003; Harley et al., 2005). As illustrated in the first and second sets of plots in Fig. 2, most of the tendencies in carbonaceous compositions were due to fluctuations in OC contents. Slight deviations were detected in day-of-week average concentrations of OC and EC (Fig. 2). Three-day average OC varied from 11.29 ± 5.84 μg m^{-3} (Tuesday-to-Thursday) to 15.39 ± 6.51 μg m^{-3} (Sunday-to-Tuesday), and the EC contributions differed from 4.16 ± 1.55 μg m^{-3} (Tuesday-to-Thursday) to 5.06 ± 1.78 μg m^{-3} (Sunday-to-Tuesday). Similarly, three-day average OC/EC ratios were between 2.48 ± 0.83 (Wednesday-to-Friday) to 3.18 ± 0.70 (Friday-to-Sunday), and the OC_{soc} values were between 65.56 ± 9.28% (Wednesday-to-Friday) to 72.22 ± 4.82% (Friday-to-Sunday). Although there were subtle changes in carbonaceous compositions, no statistically significant differences ($p < 0.05$) were found in the three-day average samples. This can be simply explained by the comparatively long sampling period of three days (i.e., 72 h). To investigate the impacts of the day-of-week trends on carbonaceous contents, all of the data were separated into two sets: a weekday group (i.e., the average of Monday to Wednesday, Tuesday to Thursday, and Wednesday to Friday) and a weekend group (i.e., Friday to Sunday). After rearranging the variables into the two data sets, the OC/EC ratios and OC_{soc} of the weekend group were statistically larger ($p < 0.05$) than those of the weekday group. Thus it seems reasonable to assume that driving patterns were the most important factor governing $PM_{2.5}$-bound carbonaceous contents in Bangkok and surpassed other factors, such as meteorology and emission source strength.

Further attempts to analyse the day-of-week trends were conducted by separating the OC/EC ratios and OC_{soc} into four different quartiles based on their values (see Table 2). The relatively low OC/EC ratios were observed during the Tuesday-to-Thursday (25% in Q1) and Wednesday-to-Friday sampling periods (25% in Q1). Likewise, comparatively low OC_{soc} values were detected in the Tuesday-to-Thursday (20.8% in Q1), Wednesday-to-Friday (20.8% in Q1), and Thursday-to-Saturday (20.8% in Q1) monitoring intervals. These findings underline the prominence of vehicular emissions as a key factor controlling black carbon content and SOC formation in the urban atmosphere during the week. The fact that 26.1% of OC_{soc} in Q4 were observed in the Saturday-to-Monday samples indicates that the low traffic densities might have allowed long-range transport of fine particles (i.e., higher contents of OC_{soc}) that approached the city during the weekend.

As noticeably seen in Fig. 3, some distinguishing features can be obtained directly from the original Gaussian distribution curve. Firstly, a sharp symmetrical bell-shape curve was found for the EC samples in comparison to those of OC and TC. Because the detected values of EC are more concentrated in the middle than in the tails, it is reasonable to assume that this is the result of less spatial variance of EC in the background air mass, which was much less affected by atmospheric photooxidation, thermal degradation, homogeneous and/or heterogeneous chemical reactions. This can also be inferred as a consequence of a prevalent contribution of vehicular exhaust on the EC levels, particularly in the urban atmosphere. On the contrary, TC and OC show exceedingly broad peaks with flat tops between 18 μg m^{-3} and 13 μg m^{-3}, respectively, which underlines the higher degree of variance than for EC. These flat symmetrical distribution curves also reveal that OC was more sensitive to meteorology and thus displays a broader peak than EC. This was in good agreement with the comparatively high percentage contribution of OC_{soc} observed at CHAOS (68.9 ± 7.22%), which supports the idea that OC appears to be more susceptible to meteorology than EC.

Table 2
The OC/EC ratios and OC_{soc} in $PM_{2.5}$ coupled with its percentage contributions at CHAOS in four different quartiles based on their values.

	OC/EC − 1[a]	OC/EC − 2[b]	OC/EC − 3[c]	OC/EC − 4[d]	OC_{soc} − 1[a]	OC_{soc} − 2[b]	OC_{soc} − 3[c]	OC_{soc} − 4[d]
Monday–Wednesday	1	5	5	1	2	6	1	3
Tuesday–Thursday	6	2	2	5	5	3	5	2
Wednesday–Friday	6	3	1	3	5	3	2	3
Thursday–Saturday	4	6	2	2	5	2	4	2
Friday–Sunday	2	3	4	5	2	5	3	4
Saturday–Monday	4	2	5	4	3	3	4	6
Sunday–Tuesday	1	2	4	3	2	1	4	3
Total count	24	23	23	23	24	23	23	23
Percentage contribution								
Monday–Wednesday	4.2	21.7	21.7	4.3	8.3	26.1	4.3	13.0
Tuesday–Thursday	25.0	8.7	8.7	21.7	20.8	13.0	21.7	8.7
Wednesday–Friday	25.0	13.0	4.3	13.0	20.8	13.0	8.7	13.0
Thursday–Saturday	16.7	26.1	8.7	8.7	20.8	8.7	17.4	8.7
Friday–Sunday	8.3	13.0	17.4	21.7	8.3	21.7	13.0	17.4
Saturday–Monday	16.7	8.7	21.7	17.4	12.5	13.0	17.4	26.1
Sunday–Tuesday	4.2	8.7	17.4	13.0	8.3	4.3	17.4	13.0
Total %	100	100	100	100	100	100	100	100

[a] First quartile (Q1).
[b] Second quartile (Q2).
[c] Third quartile (Q3).
[d] Fourth quartile (Q4).

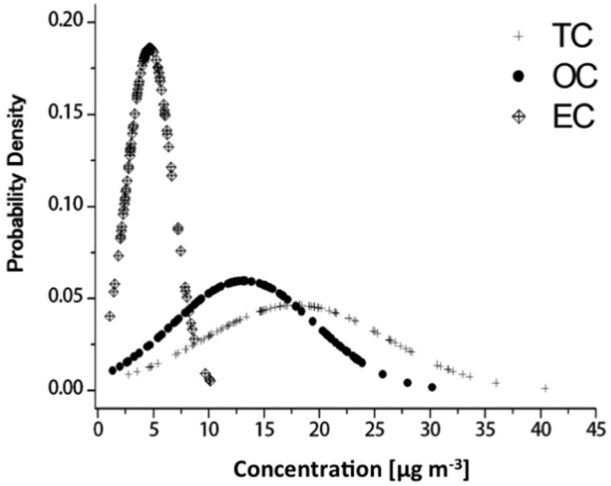

Fig. 3. Probability distribution of TC, OC and EC in $PM_{2.5}$ collected at CHAOS.

3.3. Autocorrelation of $PM_{2.5}$ and carbonaceous compositions

Generally, a correlogram is a mathematical tool for investigating randomness in a data set, which can be determined by calculating autocorrelations for data values at fluctuating time lags as described in Eqs. (3)–(5). In the case of random time series, such correlograms should be near zero for any and all time-lag intervals. In other words, the autocorrelation function should drop suddenly from 1 at zero lag to nearly zero at lags equal to one or larger. If non-random, then one or more of the correlograms will be significantly non-zero. Despite some differences in variance found in some atmospheric parameters, the time series approach employing autocorrelation plots showed a fairly strong sinusoidal wave in $PM_{2.5}$, TC, OC, and EC, as displayed in the correlograms (see Fig. 4). Because all of the autocorrelation plots showed sinusoidal wave patterns, it is reasonable to state that the stochastic oscillations of the atmospheric parameters were not random. It is also crucial to stress that the majority of the R_h values of all atmospheric parameters were lower than the confidence bands (see Fig. 4). This reveals a higher degree of impacts that may be governed by the periodic component (i.e., the day-of-week trend) rather than other confounding factors (i.e., photo degradation, thermo degradation, heterogeneous and homogeneous chemical reactions, and meteorology). In addition, the similarities of the autocorrelation plots between TC and OC were in good agreement with the comparatively high percentage contribution of OC_{soc} previously mentioned in Section 3.1, which indicates that $PM_{2.5}$-bound carbonaceous compositions in Bangkok were dominated by secondary organic carbon.

3.4. Simple linear regression analysis (SLRA)

As previously mentioned in Section 3.1, the comparatively high OC to EC ratios at CHAOS were possibly associated with high OC rather than low EC values. There are three probable reasons: anthropogenic emissions, biogenic emissions, and long-range transport of carbonaceous compositions from outside of Bangkok. Because previous studies have highlighted the importance of traffic emissions on air quality in Bangkok (Pongpiachan, 2013; Pongpiachan et al., 2013a,2013b, 2014b), biogenic emissions of carbonaceous aerosols were not likely a major source of the measured high OC contents at CHAOS. The comparative contributions of long-range transport and local biogenic emissions were investigated using a simple linear regression analysis between OC and EC contents. If the majority of the particulate OC around CHAOS was influenced by biogenic emissions, the *R*-value of OC and EC should be low because EC are mainly released from transportation. On the contrary, if the *R*-values of OC and EC are high, it can be reasonably

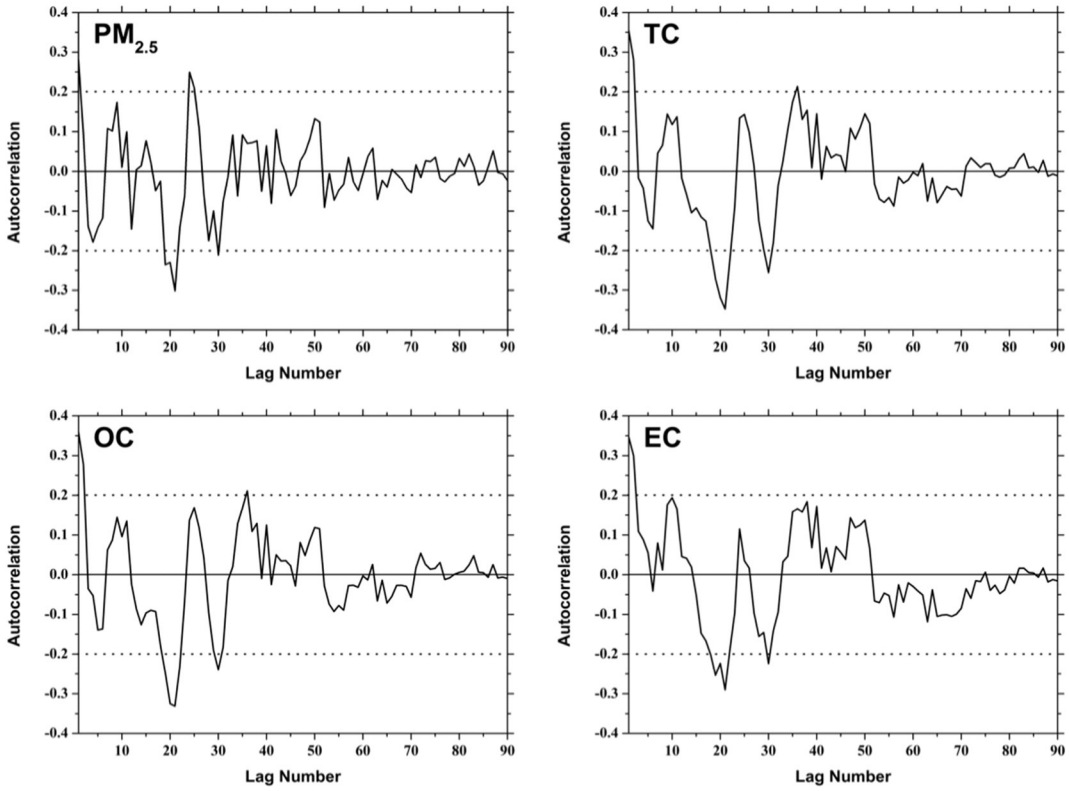

Fig. 4. Plots of autocorrelation using contents of TC, OC and EC in $PM_{2.5}$ collected at CHAOS.

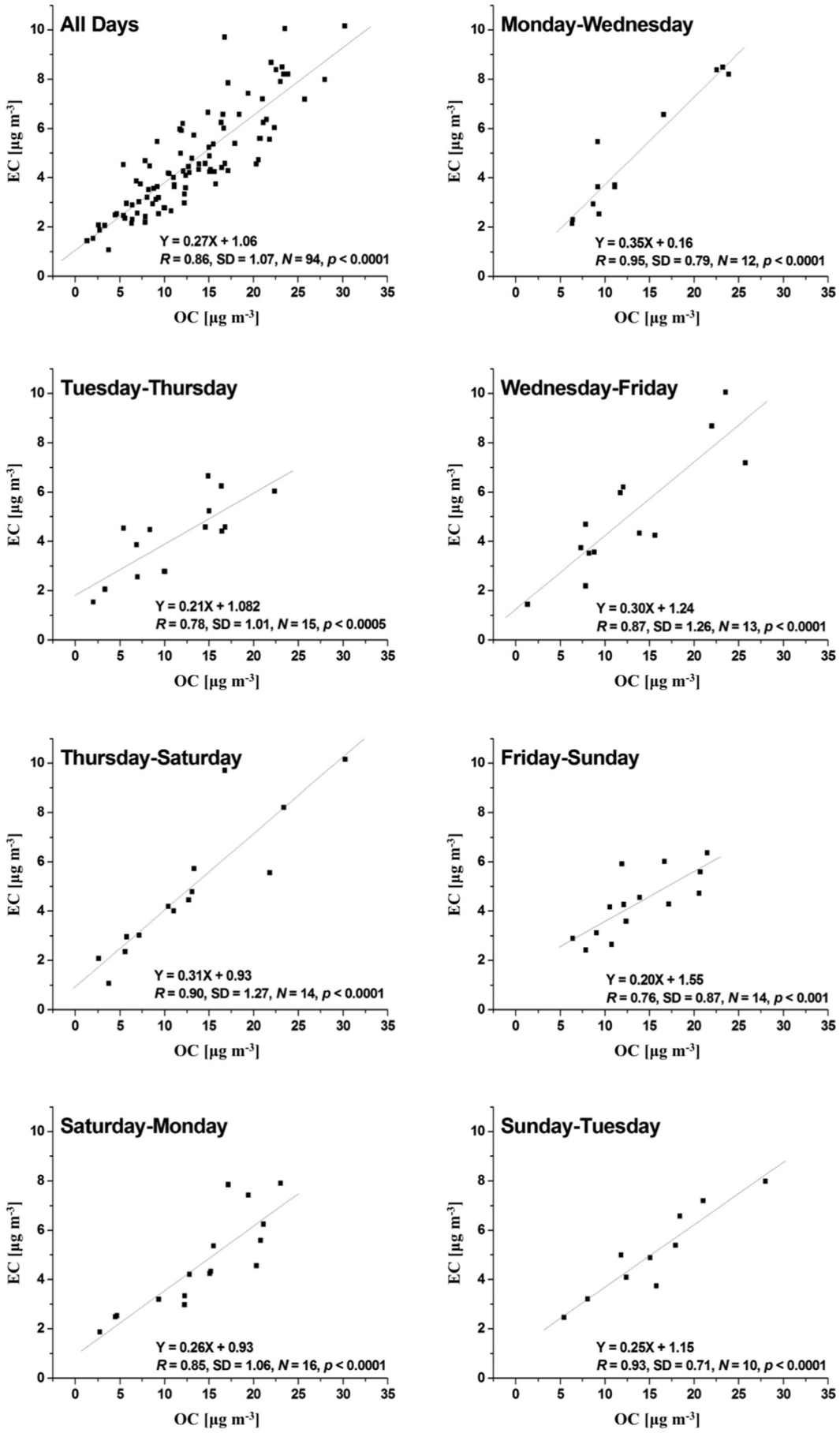

Fig. 5. Linear regression analysis of OC and EC observed in CHAOS.

concluded that both are emitted simultaneously from a single source, namely transportation (Chen et al., 2012).

To test this hypothesis and to assess the impact of the day-of-week trend to carbonaceous composition levels at CHAOS, a linear regression between the OC and EC contents in each sampling interval was applied (See Fig. 5). In general, a high R-value ($R = 0.86$) coupled with a lower p-value ($p < 0.0001$) was found in all of the sampling periods, which suggests a single dominant emission source (mostly traffic-related) in Bangkok. This finding is in good agreement with a previous study conducted in the southern China coastal area at Xiamen, which found a strong positive correlation ($R = 0.91$) between OC and EC during the winter monsoon season as a consequence of prevailing vehicular exhausts (Chen et al., 2012). On the contrary, the lowest R-value ($R = 0.76$, $p < 0.0001$) was detected in the Friday-to-Sunday samples. These results reflect a comparatively smaller contribution of mobile emissions on carbonaceous content during the weekend, which was consistent with the relatively high OC_{soc} ($72.22 \pm 4.82\%$) observed from Friday-to-Sunday (see Section 3.2). The highest R-value ($R = 0.95$, $p < 0.0001$) observed in the Monday-to-Wednesday samples indicates that both OC and EC were released simultaneously and thus underlines the importance of transportation on carbonaceous compositions during working days.

3.5. Hierarchical cluster analysis (HCA)

To obtain more information on the origins of the carbonaceous compositions collected at CHAOS, HCA was conducted on the eight variables ($n = 94$), which were TC, OC, EC, buses, Tuktuks, trucks, pick-ups/vans, and cars. The cluster analysis revealed the presence of three different groups, as displayed in the dendrogram in Fig. 6. The first cluster consisted of TC, OC, EC, buses, Tuktuks, and trucks. Because the majority of buses and trucks are powered by diesel engines, this cluster clearly indicates that heavy-duty vehicles (HDVs) appear to have been responsible for the increase of ambient carbonaceous compositions. These findings are consistent with previous studies of measurements of particulate matter from on-road vehicles and inside a tunnel, which highlighted that diesel engines had higher emission rates than did gasoline and LPG engines for most carbonaceous fractions (Cheng et al., 2010; He et al., 2006). The second sub-cluster was composed of pick-ups/vans, which is indicative of a mixing of the three types of fuel, namely "diesel", "gasohol", and "benzene". Gasohol is a mixture of gasoline and ethanol, which is an alternative fuel to 100% gasoline and helps lessen the consumption of gasoline, and has been on the market since 2001 in Thailand. (See Fig. 6.)

The third cluster contained only personal cars, for which gasohol is the main fuel. On January 1, 2013, the Thailand government abandoned the sale of regular octane 91 gasoline to promote gasohol usage. As a consequence, ethanol consumption rapidly rose from 1.3 million L day^{-1} in 2012 to 2.0 million L day^{-1} in 2014. The Department of Alternative Energy Development and Efficiency, Ministry of Energy, launched the new energy policy with a target to enhance ethanol usage, primarily of E20 and E85, to 3.0 million L day^{-1} in 2015 and to 9.9 million L day^{-1} in 2021. This will inevitably affect the emission factors of OC and EC from vehicular exhausts and thus the atmospheric contents of carbonaceous aerosols in the near future. As a consequence, it is reasonable to conclude that the proximity in the dendrogram of vehicle type vs. carbonaceous compositions is directly related to the fuel and engine types. In addition, driving patterns coupled with road conditions can dramatically change the emission factors of OC and EC and thus need to be considered in future work.

3.6. Principal component analysis (PCA)

Generally, PCA can be applied as a multivariate statistical tool to reduce a set of original variables (i.e., $PM_{2.5}$-bound carbonaceous compositions and vehicle types) and to extract a small number of latent factors (i.e., principal components, PCs), to investigate associations among the measured parameters. The data that were accepted for investigation were organized into a matrix, where each column was a parameter component and the samples were in the rows. The data matrices were analysed using PCA, which permitted the reviewed data to be further assessed and plotted in three dimensions. In this study, the principal component patterns for Varimax rotated components were composed of three components, namely PC1 (36.9%), PC2 (27.2%) and PC3 (17.2%), which accounted for 81.3% of the total variance.

The clearest 3D plot features displayed in Fig. 7 are (*i*) the TC, OC, EC, and pick-ups/vans are grouped together, (*ii*) Tuktuks and buses are highly separated from the carbonaceous compositions and pick-ups/vans, and (*iii*) the cars and trucks are closer to the TC, OC, and EC group than Tuktuks and buses. While the dendrogram shows the close proximities of carbonaceous composition groups with buses, Tuktuks, and trucks (see Section 3.5), the PCA 3D plots show the strong associations between carbonaceous aerosols and pick-ups/vans. This discrepancy may merely reflect a difference in the statistical analogy between PCA and HCA. While PCA is a multivariate statistical technique used for reducing a set of elements by selecting the attributes with the most variation, HCA is an unsupervised learning method to find groups of similarities based on attribute values. Despite the differences in these two statistical tools, the strong influence of diesel vehicles (e.g., buses, pick-ups/vans, trucks) on $PM_{2.5}$-bounded carbonaceous particles is undoubtedly obvious. Because the majority of pick-ups/vans consume

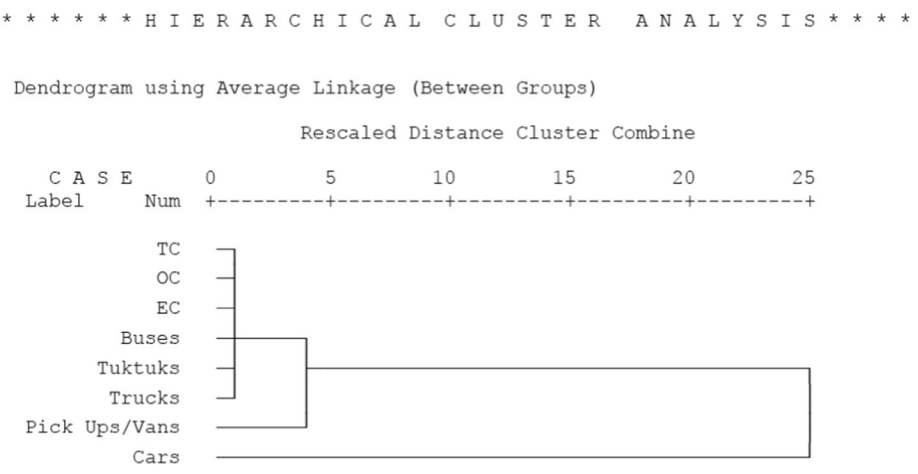

Fig. 6. Dendrogram of hierarchical cluster analysis using average linkage (between groups).

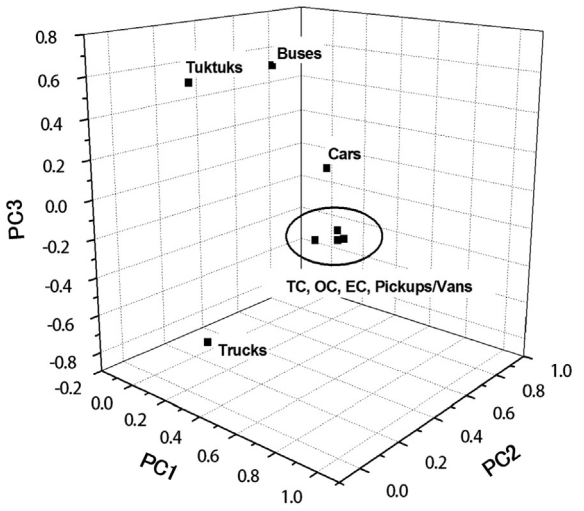

Fig. 7. Three-dimensional plots of principal components of TC, OC, EC, pickups/vans, trucks, cars, buses and Tuktuks.

diesel fuels, it appears reasonable to ascribe the strong affinity between carbonaceous compositions and pick-up/vans as a consequence of the diesel engine combustion process. This interpretation is also supported by the 3D proximity of cars and trucks with the carbonaceous aerosols. In addition, it is also noteworthy that the air sample collections in the tunnel prove that diesel engine emissions contribute most of the carbonaceous compositions in the urban atmosphere (Cheng et al., 2010; He et al., 2006).

4. Conclusions

This study attempts to conduct the time series analysis based on the long term monitoring of $PM_{2.5}$-bounded OC/EC collected at ONE monitoring site. According to our best knowledge, this is the only long term monitoring data of $PM_{2.5}$-bounded carbonaceous fractions in Thailand. Since this study use the 48 h sampling period data, it appears reasonable to interpret that the impacts of local thermal circulation are minor of importance. The analysis of day-of-week trends of OC/EC ratios, the time series results and OC_{soc} reveal that vehicle fleets are the most influential factors governing the atmospheric content of $PM_{2.5}$-bound carbonaceous compositions. Although traffic emissions played an important role in controlling carbonaceous aerosols during the week, multiple types of emission sources, including the long-range transport, appear to have been the main contributors on weekends. Irrespective of some discrepancies that occurred from the application of HCA and PCA to investigate the relationship between carbonaceous compositions and vehicle types, the overwhelming contribution from diesel emissions on the $PM_{2.5}$-bound TC, OC, and EC is unquestionably evident. This leads to greater public health concerns of urban air quality. Overall, a variety of emission control strategies for different categories of in-use HDVs, coupled with multiple effective solutions for most fleets in both weekday and weekend periods, will be required to improve air quality in Bangkok.

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