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Carbon composition of PM_{10} and $PM_{2.5}$ in Bangkok ambient air from a city center sampling site

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Abstract

PM_{10} and $PM_{2.5}$ concentrations can adversely affect the respiratory and cardiac health of humans and also reduce visibility. Black carbon (BC) aerosols absorb solar radiation and have been shown to be the second largest contributor to global warming after greenhouse gases. Organic carbon (OC) and elemental carbon (EC) composition differs for different emission sources, and this difference is useful in identifying and controlling both $PM_{2.5}$ and BC, the two major causes of air pollution and global warming. The objectives of this study were to measure the carbon composition of aerosols and to find relationships among the different constituents that can be used to identify the emission sources of PM_{10}, $PM_{2.5}$ and BC in the ambient air of the Bangkok city center at Chulalongkorn University. PM_{10} and $PM_{2.5}$ were collected with a dichotomous air sampler located on the roof of the five-story Environmental Engineering building. The samples were collected for 24 hours every 6 days in both dry and wet seasons from December, 2011, to July, 2012. OC and EC were analyzed with thermal-optical methods. Concentrations of BC were measured with a microaethalometer for 15 days in the dry season. The results showed that in the dry season, the average PM_{10} and $PM_{2.5}$ concentrations were 80 $\mu g/m^3$ and 48 $\mu g/m^3$, respectively. The average ratio of $PM_{2.5}$ to PM_{10} concentrations was 0.60. In the wet season, PM_{10} and $PM_{2.5}$ concentrations were less than in the dry season: 45 $\mu g/m^3$ and 23 $\mu g/m^3$, respectively. The average ratio of $PM_{2.5}$ to PM_{10} concentrations was 0.51. The PM_{10} and $PM_{2.5}$ concentrations in the wet season were approximately half of the concentrations in the dry season. BC concentrations ranged from 2.33 – 5.50 $\mu g/m^3$, with a mean of 3.64 $\mu g/m^3$. A diurnal variation was observed, with the highest values occurring on weekdays at approximately 6–9 a.m. and in the evening, which correspond to peak traffic in the morning and afternoon. In the dry season, the average concentration of EC in the $PM_{2.5}$ was 3.04 $\mu g/m^3$. The ratio of total carbon (TC), OC and EC concentrations to the $PM_{2.5}$ concentration were 0.35, 0.29 and 0.06, respectively. The average ratio of OC to EC concentration in $PM_{2.5}$ was 4.52. The average ratio of the Char-EC to Soot-EC concentration in $PM_{2.5}$ was 1.70. Major fractions of eight carbon fractions (OC1, OC2, OC3, OC4, EC1, EC2, EC3 and optically detected pyrolyzed carbon (OP)) were EC1- and OP-enriched in motor vehicle exhaust. We conclude that controlling motor vehicle emissions is the most important factor in controlling both particulate matter and black carbon in the Bangkok city center ambient air.

Keywords: *PM_{10}, $PM_{2.5}$, Black carbon (BC), Organic carbon (OC), Elemental carbon (EC), Char-EC, Soot-EC*

บทคัดย่อ

ฝุ่นขนาดเล็กกว่า 10 ไมครอน (PM_{10}) และ ฝุ่นขนาดเล็กกว่า 2.5 ไมครอน ($PM_{2.5}$) สามารถส่งผลกระทบต่อสุขภาพของมนุษย์ทั้งระบบทางเดินหายใจ และโรคหัวใจ และยังลดประสิทธิภาพในการมองเห็น แบล็กคาร์บอน (BC) มีผลต่อการดูดซึมรังสีความร้อน และเป็นแหล่งกำเนิดสำคัญลำดับสองในการทำให้โลกร้อนรองจากก๊าซเรือนกระจก ชนิดของคาร์บอนได้แก่ คาร์บอนอินทรีย์ (Organic Carbon - OC) และธาตุคาร์บอน (Elemental Carbon - EC) มีแหล่งกำเนิดแตกต่างกัน ดังนั้นองค์ประกอบคาร์บอนในฝุ่นละอองสามารถใช้บ่งชี้แหล่งกำเนิด และเป็นแนวทางเพื่อควบคุมทั้งฝุ่นขนาดเล็ก และแบล็กคาร์บอนซึ่งเป็นปัญหาหลักในการเกิดมลพิษอากาศและภาวะโลกร้อน วัตถุประสงค์ของการศึกษานี้คือตรวจวัดองค์ประกอบคาร์บอนในฝุ่นละอองในบรรยากาศ หาความสัมพันธ์และความแตกต่างขององค์ประกอบเพื่อใช้บ่งชี้แหล่งกำเนิดของ PM_{10} $PM_{2.5}$ และ BC ในบรรยากาศบริเวณใจกลางกรุงเทพมหานครที่จุฬาลงกรณ์มหาวิทยาลัย โดยใช้เครื่องเก็บตัวอย่างฝุ่นสองขนาด (Dichotomous sampler) ตั้งเครื่องมือที่ดาดฟ้าตึกวิศวกรรมสิ่งแวดล้อม 5 ชั้น เก็บตัวอย่างฝุ่น เฉลี่ย 24 ชั่วโมง ทุกๆ 6 วัน ทั้งในฤดูแล้ง และฤดูฝน ตั้งแต่เดือนธันวาคม 2554 - กรกฎาคม 2555 วิเคราะห์ EC และ OC ด้วยวิธีความร้อนและแสง (thermal-optical methods) ตรวจวัด BC ด้วยเครื่อง Micro Aethalometer เป็นเวลา 15 วันในฤดูแล้ง ผลการศึกษาพบว่า ในฤดูแล้งความเข้มข้นของ PM_{10} และ $PM_{2.5}$ มีค่าเฉลี่ย 80 มคก./ลบ.ม. และ 48 มคก./ลบ.ม. ตามลำดับ สัดส่วนความเข้มข้น $PM_{2.5}/PM_{10}$ มีค่าเฉลี่ย 0.60 ในฤดูฝนความเข้มข้นของ PM_{10} และ $PM_{2.5}$ มีค่าเฉลี่ย 45 มคก./ลบ.ม. และ 23 มคก./ลบ.ม. ตามลำดับ สัดส่วนความเข้มข้น $PM_{2.5}/PM_{10}$ มีค่าเฉลี่ย 0.51 ความเข้มข้น PM_{10} และ $PM_{2.5}$ ในฤดูฝนมีค่าต่ำกว่าฤดูแล้งประมาณครึ่งหนึ่ง ความเข้มข้นของ BC เฉลี่ยรายชั่วโมงอยู่ในช่วง 2.33 ถึง 5.50 มคก./ลบ.ม. มีค่าเฉลี่ย 3.64 มคก./ลบ.ม. ในระหว่างวันทำงานความเข้มข้นของแบล็กคาร์บอนเฉลี่ยรายชั่วโมงสูงสุดอยู่ในช่วง 6.00 – 9.00 น. และเวลาเย็น เนื่องจากการจราจรเร่งด่วนในช่วงเช้าและเย็น ในฤดูแล้งความเข้มข้น EC ใน PM_{10} และ $PM_{2.5}$ มีค่าเฉลี่ย 11.98 มคก./ลบ.ม. และ 3.04 มคก./ลบ.ม. ตามลำดับ ความเข้มข้นของ Total Carbon (TC) OC และ EC ใน $PM_{2.5}$ ต่อความเข้มข้นของ $PM_{2.5}$ มีค่าเท่ากับ 0.35

0.29 และ 0.06 ตามลำดับ สัดส่วนระหว่าง OC/EC ใน PM_{10} และ $PM_{2.5}$ มีค่าเฉลี่ย 4.91 และ 4.52 ตามลำดับ สัดส่วนความเข้มข้นระหว่าง char-EC: soot-EC ใน PM10 และ PM2.5 มีค่าเฉลี่ย 0.96 และ1.70 ตามลำดับ ความเข้มข้นของคาร์บอนทั้ง 8 ประเภท (OC1 OC2 OC3 OC4 EC1 EC2 EC3 และ OP) ที่พบสูงสุดได้แก่ EC1 และ OP บ่งชี้ถึงแหล่งกำเนิดมลพิษมาจากยานพาหนะ จึงสรุปได้ว่าการควบคุมไอเสียจากยานพาหนะมีความสำคัญสูงสุดในการควบคุมฝุ่นละอองและแบล็กคาร์บอนในบรรยากาศกลางเมืองกรุงเทพมหานคร

คำสำคัญ: *PM_{10}, $PM_{2.5}$, แบล็กคาร์บอน (BC), คาร์บอนอินทรีย์ (OC), ธาตุคาร์บอน (EC), Char-EC, Soot-EC*

1. Introduction

Carbon, one of the elements in atmospheric particulate matter, accounts for 20-60% of the $PM_{2.5}$ concentration (Gu, Bai, Liu, Wu, Xie, Li, Dong, & Zhang, 2010) and exists mainly in the forms of organic carbon (OC) and elemental carbon (EC, or black carbon, BC). Recently, atmospheric BC has received considerable attention because of its significant influence on climate change, as well as its adverse effects on human health. Reducing BC aerosols can reduce global warming and benefit human health (Suwattiga, 2011). BC is emitted from incomplete combustion processes, such as burning fossil fuels, biofuels and biomass. Most of the BC aerosols (90%) reside in the $PM_{2.5}$ fraction (Viidanoja, Sillanpaa, Laakia, Kerminen, Hillamo, Aarnio, & Koskentalo, 2002), and BC constitutes 5-15% of the $PM_{2.5}$ in urban air. EC is generally subdivided into char and soot. Soot is composed of submicron particles formed from the condensation of hydrocarbon radicals at high temperatures (>600 ºC). Char retains the morphology of the source material with particle diameters ranging from 1 to 100 μm. Because the different classes of EC have different chemical and physical properties, their optical light-absorbing properties differ. The ratio of char and soot would help us better understand their environmental and climatic impacts (Han, Cao, Lee, Ho, & An, 2009). Knowledge of carbon composition of aerosols is also useful in identifying source and control measures for both $PM_{2.5}$ and BC, the two major causes of air pollution and global warming.

2. Objectives

The objectives of this study were to measure the carbon composition of aerosols to find relationships among the different constituents that could be used to identify the emission sources of PM_{10}, $PM_{2.5}$ and BC in the ambient air of Bangkok.

3. Materials and methods

PM_{10} and $PM_{2.5}$ were collected with a dichotomous air sampler at Chulalongkorn University on the rooftop of the 5-story environmental engineering building located in the Bangkok city center (Figure 1). The high elevation of the sampler allowed for a better mixing of Bangkok background air because the sampler was some distance from street-level emission sources. The height is also relevant to occupants of high-rise buildings in the area in terms of small particle exposure. The samples were collected on quartz fiber filters for 24 hours every 6 days in both dry and wet seasons from December, 2011, to July, 2012. The quartz filters were pre-heated in a furnace at 550 °C for six hours before sampling to remove the residual carbon. Before and after sampling, the filters were conditioned in desiccators for 48 hours and then weighed on an electronic balance with a ±0.01 mg sensitivity to determine the PM mass. Each filter was weighed at least 3 times before and after sampling. After weighing, the samples were placed in a refrigerator at -4 °C until analysis. In this study, a total of 29 $PM_{2.5}$ and 29 PM_{10} samples were collected during the sampling periods.

Organic carbon (OC), elemental carbon (EC), Char-EC, and Soot-EC were analyzed with thermal-optical methods following the IMPROVE (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Chow, Watson, Chen, Chang, Robinson, Trimble, & Kohl, 2007). In the analysis procedure, a 0.5 cm^2 punch from the filter was analyzed, and the four OC fractions (OC1, OC2, OC3, and OC4) were obtained at 120 °C, 250 °C, 450 °C, and 550 °C in a He atmosphere. The pyrolyzed carbon fraction (OP) was determined when a reflected laser light attained its original intensity after O_2 was added to the experimental atmosphere. Three EC fractions (EC1, EC2, and EC3) were obtained at 550 °C, 700 °C, and 800 °C, respectively, in a 2% O_2/98% He atmosphere. Based on the IMPROVE protocol, OC is defined as OC1 + OC2 + OC3 + OC4 + OP and EC is defined as EC1 + EC2 + EC3 - OP. TC is the sum of OC and EC. Han, Cao, Chow, Watson, Fung, Jin, Liu, and An (2007) defined Char-EC as EC1-OP and Soot-EC as EC2+EC3.

The BC concentration was continuously measured with a microaethalometer (Figure 2) for 15 days in the dry season.

4. Results and discussion

4.1 Concentrations of PM_{10} and $PM_{2.5}$ and ratios of PM_{10} to $PM_{2.5}$

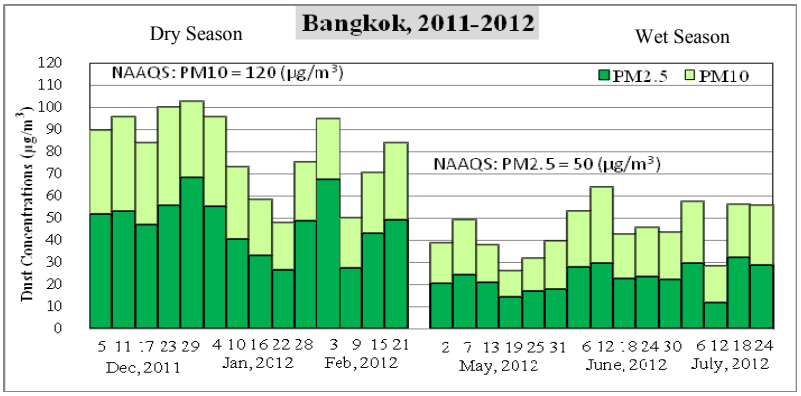
The dry season and the wet season can be differentiated by comparing monthly rainfall intensity to annual average rainfall intensity. For the purpose of this study, the dry season was from December to February, and the wet season was from May to July (Figure 3).

In the dry season, 24-hr PM_{10} concentrations were between 48-113 $\mu g/m^3$ (National Ambient Air Quality Standards - NAAQS is 120 $\mu g/m^3$). The 24-hr $PM_{2.5}$ concentrations were 27-69 $\mu g/m^3$ (the NAAQS is 50 $\mu g/m^3$). The average dry season PM_{10} and $PM_{2.5}$ concentrations were 80 $\mu g/m^3$ and 48 $\mu g/m^3$, respectively. In the wet season, the 24-hr PM_{10} concentration range was 26 - 64 $\mu g/m^3$. The 24-hr $PM_{2.5}$ concentrations were between 12 - 32 $\mu g/m^3$. The average wet season PM_{10} and $PM_{2.5}$ concentrations were 45 $\mu g/m^3$ and 23 $\mu g/m^3$, respectively (Figure 3). The PM_{10} and $PM_{2.5}$ concentrations in the wet season were approximately half of the concentrations observed in the dry season. All concentrations of PM_{10} at the study site were within NAAQS, but concentrations of $PM_{2.5}$ in the dry season (6 samples) exceeded NAAQS.

The average ratio of $PM_{2.5}$ to PM_{10} concentrations in the dry season and the wet season were 0.60 and 0.51, respectively. Particles smaller than 2.5 microns accounted for slightly more than half of the PM_{10} (Table 1). This ratio is similar to the average ratio of $PM_{2.5}$ to PM_{10} at Dindaeng Station (0.67 and 0.59). The Pollution Control Department's Dindaeng Station is a roadside station that records a greater proportion of fine particles from traffic exhaust. The correlation analysis found a good correlation between $PM_{2.5}$ concentrations at Dindaeng Station and Chulalongkorn University in both the dry and wet seasons (r = 0.72 and r = 0.75, respectively).

Figure 1 Sampling Site at Chulalongkorn University

Figure 2 Microaethalometer



Rainfall intensity
Wet season 121.9-196.4 mm
Dry season 0.7-64.9 mm
Source: Meteorological Department

Figure 3 PM_{10} and $PM_{2.5}$ concentrations in the dry and wet seasons

Table 1 Ratio of $PM_{2.5}$ to PM_{10} at Dindaeng Station and Chulalongkorn University

		Dry season	Wet season
Ratio of $PM_{2.5}$ to PM_{10}	Dindaeng Station	0.67	0.59
	Chulalongkorn University	0.60	0.51
Correlation (r) between Dindaeng and Chula	PM10	0.63	0.65
	PM2.5	0.72	0.75

Source: Dindaeng Station data from PCD

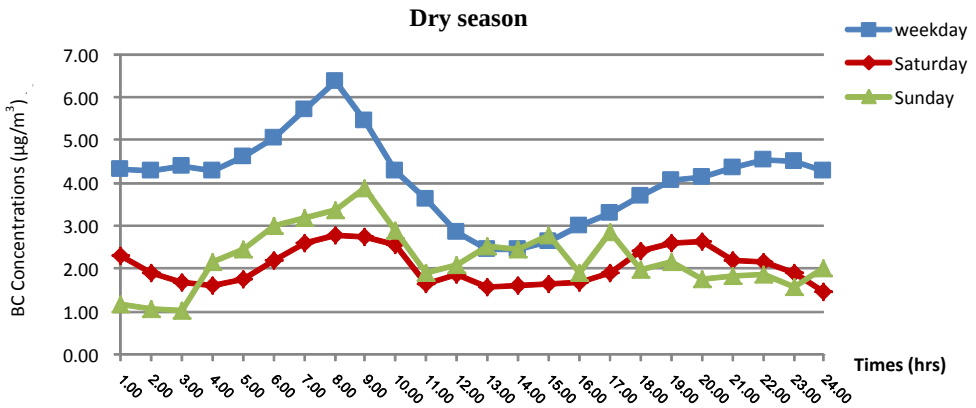


Figure 4 The average hourly diurnal variation in BC concentration in the dry season

4.2 Diurnal variation of Black carbon concentrations

The average 1-hr BC concentrations in the dry season ranged from 2.33 to 5.50 µg/m^3, with a mean of 3.64 µg/m^3. During weekdays (Monday to Friday, 11 samples), a diurnal variation was observed, with the highest values occurring at approximately 6–9 a.m. and another peak in the evening (Figure 4); conversely, on Saturday (2 samples), the peak observed in the morning was lower than those on weekdays and was followed by low concentrations during the rest of the day. On Sunday (2 samples), pollutant concentrations were low throughout the day, corresponding to the traffic on Sunday.

4.3 Carbon compositions

The carbon compositions are summarized in Table 2 for the dry season only because the carbon concentrations in the wet season were too low to be reliable. The average concentration of EC in $PM_{2.5}$ was 3.04 µg/m^3. The average concentration of OC in $PM_{2.5}$ was 14.00 µg/m^3. The ratios of TC, OC and EC concentrations to the $PM_{2.5}$ concentration were 0.35, 0.29 and 0.06, respectively. The total carbon composition of aerosols could be estimated to be approximately 35% of the $PM_{2.5}$ concentrations.

4.4 Source identification from OC/EC ratios and Char-EC/Soot-EC ratios

The average ratios of OC to EC and Char-EC to Soot-EC have been used to identify the source of carbonaceous aerosols, as shown in Table 3.

At this study site, the average ratio of OC to EC concentration in $PM_{2.5}$ was 4.52 (Table 4), indicating that the sources were gasoline and LPG exhaust. The average ratio of Char-EC to Soot-EC concentration in $PM_{2.5}$ was 1.70, indicating that the sources were motor vehicle emissions.

Table 2 Carbon composition and the average ratios of TC, OC and EC to particulate matter in the dry season

Period	Dust types		Concentrations (μg/m ^{3})	TC (μg/m ^{3})	TC/PM	OC (μg/m^3)	OC/PM	EC (μg/m ^{3})	EC/PM
Dry season 5 Dec. 2011- 27 Feb. 2012 (n=14)	$PM_{2.5}$	Min	26.85	6.49	0.23	5.13	0.19	1.36	0.05
		Max	68.52	31.64	0.64	27.17	0.55	4.52	0.09
		Avg	47.71	17.04	0.35	14.00	0.29	3.04	0.06

Table 3 Source identification using OC/EC and Char-EC/Soot-EC ratios

Sources	OC/EC	Char-EC /Soot-EC
Gasoline and LPG exhaust	1.0 – 4.0 (Schauer et al., 1999, 2002)	1.0 -2.0 (Chow et al., 2004)
Diesel exhaust	< 1 (Schauer et al., 1999, 2002)	1.0 -2.0 (Chow et al., 2004)
Biomass combustion	7.0 – 8.0 (Zhang et al., 2007)	2.0 - 5.0 (Chen et al., 2007)
Wood combustion	16.8 – 40.0 (Schauer et al., 2002)	-
Residential coal combustion	2.5 – 10.5 (Chen et al., 2006)	1.5 – 3.0 (Cao et al., 2005)
Residential cooking produced	32.9 – 81.9 (He et al., 2004)	2.0 - 6.0 (Chow et al., 2004)

Table 4 The average ratios of OC/EC and Char-EC/Soot-EC in $PM_{2.5}$ and PM_{10} in the dry season

Period	Dust types		OC (μg/m^3)	EC (μg/m ^{3})	OC/EC	Char-EC (μg/m ^{3})	Soot-EC (μg/m ^{3})	CharEC/ Soot-EC
Dry season 5 Dec 2011- 27 Feb 2012 (n=14)	$PM_{2.5}$	Min	5.13	1.36	3.36	0.66	0.70	0.90
		Max	27.17	4.52	6.07	3.23	1.56	2.62
		Avg	14.00	3.04	4.52	1.94	1.11	1.70

Table 5 Source identification for eight carbon fractions

Eight Carbon Fractions	Sources	References
OC1	abundant in biomass burning	Chow et al., 2004
OC2	abundant in coal combustion	Chow et al., 2004.
OC3	abundant in gasoline and LPG exhaust	Cao et al, 2006
OC4	abundant in road dust profile	Cao et al., 2005
OP	abundant in incomplete combustion of fossil fuels, meat cooking and biomass burning	Cao et al., 2005
EC1	enriched in motor vehicle exhaust	Cao et al., 2005
EC2	enriched in diesel exhaust	Cao et al., 2006
EC3	enriched in coal combustion and motor vehicle exhaust	Yu et al., 2002

4.5 The analysis of eight carbon fractions

The abundance of eight carbon fractions in the source sample showed the specific character of the source compositions. Eight carbon fractions were used to determine the source apportionment of the carbonaceous aerosols, as shown in Table 5.

The percentage of carbon fractions in PM_{10} and $PM_{2.5}$ collected in the dry season are shown in Figure 5. The average abundances of OC1, OC2, OC3, OC4, EC1, EC2, EC3 and OP in $PM_{2.5}$TC were 0.9%, 14.8%, 14.1%, 3.8%, 34.7%, 5.3%, 0.02% and 26.3%, respectively, and the average abundances were 2.2%, 14.1%, 18.6%, 6.9%, 28.8%, 6.8%, 0.07% and 22.5%, respectively, in PM_{10}TC. The major fractions of the eight carbon fractions were EC1 and OP. The highest abundance of EC1 and the third highest abundance of OC2 indicate major contributions from motor vehicle emissions. The second highest OP fraction implied incomplete combustion of fossil fuels (Gu et al., 2010).

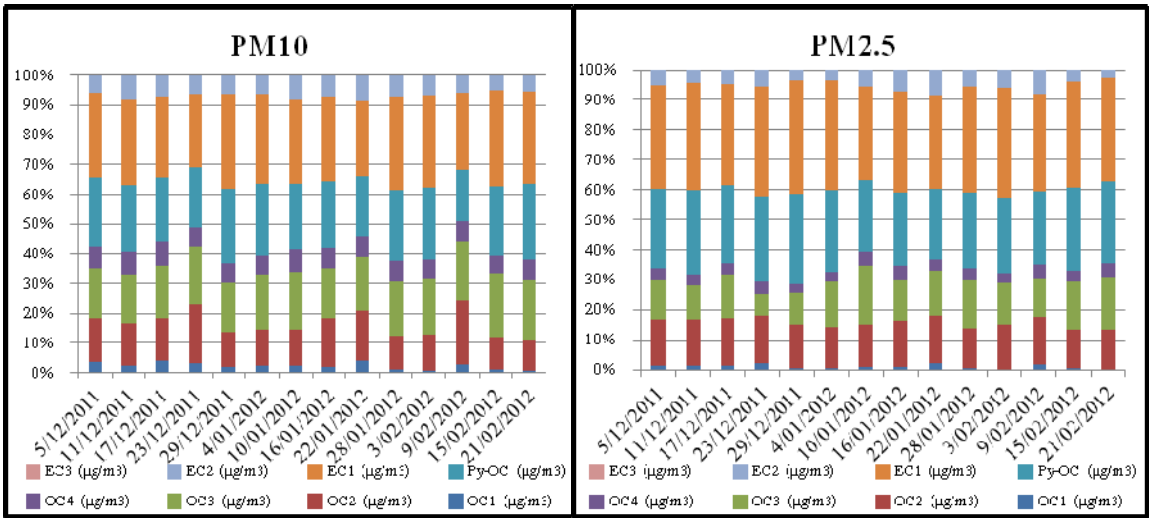


Figure 5 Percentages of eight carbon fractions in PM_{10} and $PM_{2.5}$

5. Conclusions

The $PM_{2.5}/PM_{10}$ ratios in Bangkok city center (Chulalongkorn University) averaged 0.60 and 0.51 in the dry and wet seasons, respectively. Total carbon compositions could be estimated to be approximately 35% of the $PM_{2.5}$ concentration. The average ratios of OC to EC and Char-EC to Soot-EC concentrations in PM_{10} and $PM_{2.5}$ indicate that motor vehicle emissions are the major aerosol source. The results also show that EC1 and OP are major fractions, which corresponds to motor vehicle emissions. The weekday diurnal variation of BC corresponded to peak traffic in the morning and afternoon. We conclude that controlling motor vehicle emissions is most important factor in controlling both particulate matter and black carbon in the Bangkok ambient air.

6. Acknowledgments

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