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Chemical Characterization of Carbonaceous PM_{10} in Bangkok, Thailand

[1]Siwatt Pongpiachan, [2]Shinji Kudo and [3]Kazuhiko Sekiguchi

[1]National Institute of Development Administration, Center for Research and Development of Disaster Prevention and Management, School of Social and Environmental Development, 118 Moo 3, Sereethai Road, Klong-Chan, Bangkapi, Bangkok, 10240, Thailand

[2]Center for Global Environmental Research (Global Atmospheric Chemistry Section), National Institute for Environmental Studies, Tsukuba, Japan

[3]Graduate School of Science and Engineering, Saitama University, Saitama, Japan

Corresponding Author: Siwatt Pongpiachan, National Institute of Development Administration, Center for Research and Development of Disaster Prevention and Management, School of Social and Environmental Development, 118 Moo 3, Sereethai Road, Klong-Chan, Bangkapi, Bangkok, 10240, Thailand Tel: 00 66 2 727 3113 Fax: 00 66 2 732 0276

ABSTRACT

Four Organic Carbon (OC) compositions and three Elemental Carbon (EC) fractions in PM_{10} collected monthly from February to December 2007 at eight PCD air quality observatory sites were analysed by using a DRI Model 2001. Since both OC and EC play a major role in governing the gas-particle partitioning of carcinogenic Polycyclic Aromatic Hydrocarbons (PAHs), it is therefore, important to investigate the spatial and temporal distributions of carbonaceous fractions in Bangkok. The sum of OC (ΣOC) and EC (ΣEC) collected at eight PCD sites were 71.4±19.3 μg m^{-3} and 74.9±32.7 μg m^{-3}, correspondingly. The relatively low OC/EC ratios observed in Bangkok highlight the influence of transportation sector in governing carbonaceous aerosols, particularly in heavy traffic congestion area. Three-dimensional plots of Principal Components (PCs) successfully discriminate "traffic emission" group from those of "urban residential background" group. In addition, the estimated Incremental Lifetime Particulate Exposure (ILPE) of carbonaceous compositions were constantly highest at heavy traffic congestion area in both genders with the average values of 1,130±457 and 622±252 mg for TC accumulated in male and female, respectively.

Key words: Carbonaceous compositions, PM_{10}, Bangkok, multiple linear regression analysis, principal component analysis, incremental lifetime particulate exposure

INTRODUCTION

The character of carbonaceous aerosols which are composed of OC and EC particles, has been continuously investigated during the past three decades (Dan *et al*., 2004; Duan *et al*., 2004; Ellis and Novakov, 1982; Fang *et al*., 1999; Ho *et al*., 2002; Pathak *et al*., 2011). Several concerns over their adverse impact on both human health and the climate have been raised by international scientists reflecting their importance (O'Brien and Mitchell, 2003; Repine *et al*., 2008; Shih *et al*., 2008). Recent studies have comprehensively examined the application of OC/EC ratios as indicators

of primary and secondary organic carbon (Pio *et al*., 2011; Yu *et al*., 2004; Zeng and Wang, 2011). Despite numerous studies involving the identification of OC/EC compositions in the atmospheric environment around the world (Chu, 2005; Plaza *et al*., 2006; Ram and Sarin, 2010), there is limited data regarding the character of carbonaceous aerosols in tropical countries (Pongpiachan *et al*., 2009, 2013a). Some studies have focused on the chemical composition of aerosols and their impact on the tropical/sub-tropical urban environment over the past two decades (Allena *et al*., 2004; Latha and Badarinath, 2003, 2005; Pandey *et al*., 2006; Pongpiachan, 2013a-c; Pongpiachan *et al*., 2013a-c) while others have examined the empirical estimation of Secondary Organic Carbon (SOC) formation (Grivas *et al*., 2012; Kim *et al*., 2012; Seguel *et al*., 2009; Wang *et al*., 2012). Only a few publications have presented in-depth, quantitative evidence regarding the behavior of carbonaceous aerosols in Thailand (Li *et al*., 2013; Pongpiachan *et al*., 2009, 2013a; Sahu *et al*., 2011).

However, there have been an increasing number of articles to appear in several peer-reviewed international journals over the past few years which relate to incremental lifetime cancer risk (Wiwatanadate and Liwsrisakun, 2011), inhalation exposure and lung cancer risk of ambient atmospheric PAHs (Xia *et al*., 2013) and incremental lifetime exposure to carbonaceous aerosols in southern part of Thailand (Pongpiachan *et al*., 2009). Unfortunately, there is no report related to the risk assessment of carbonaceous particles inhalation in Bangkok. Overall, the main purpose of this research was to (1) Investigate the temporal and spatial distribution of OC/EC composition in PM_{10} ateight PCD air quality observatory sites in Bangkok, (2) Empirically estimate the formation of SOC in corresponding eight different locations and (3) Calculate the incremental lifetime exposure to carbonaceous PM_{10} for Bangkokian. The monitoring period was conducted monthly from February to December 2007.

MATERIALS AND METHODS

Air sample observatory sites: Eight air sample observatory sites, namely Klongchan Housing Authority (KHA; 13°49'11.761"N 100°34'33.190"E), Nonsree High School (NHS; 13°42'28.937"N 100°32'50.443"E), Watsing High School (WHS; 13°41'3.218"N 100°26'45.554"E), Electricity Generating Authority of Thailand (EGAT; 13°43'39.205"N 100°29'11.776"E), Chokchai 4 Police Station (CPS; 13°47'33.474"N 100°35'45.879"E), Dindang Housing Authority (DHA; 13°46'59.544"N 100°32'25.618"E), Huakwang Observatory Site (HOS; 13°74'75"N 100°56'73"E) and Badindecha High School (BHS; 13°46'10.745"N 100°36'52.433"E) were selected for the investigation of carbonaceous compositions in PM_{10}. It is crucial to note that CPS, DHA, EGAT and HOS were located close to roadsides, whilst KHA, NHS, WHS and BHS were situated at the residential areas. As a consequence, "CPS, DHA, EGAT, HOS" and "KHA, NHS, WHS, BHS" can be considered as representatives of "traffic emissions" and "urban residential background", respectively. Intensive monitoring campaigns were performed at all observatory sites simultaneously at a normal weekday every first Monday of month from February to December 2007 forming a database of 85 individual air samples. Andersen high volume air sampler PM_{10} TE6001 with a flow rate of 1.132 m^3 min^{-1} was used for collecting PM_{10}. A more detailed description of the air sampling method is provided in "Compendium Method IO-2.2". Sampling of ambient air for PM_{10} using an "Andersen Dichotomous Sampler" (USEPA, 1999). The PM_{10} samples were collected on Whatmanglass microfiber filters (GFFs) for the analysis of carbonaceous compositions.

Organic Carbon (OC) and Elemental Carbon (EC): The PM_{10} mass loadings were measured gravimetrically with a Mettler Toledo AB204-S electronic microbalance (Columbus, Ohio, USA). Prior to aerosol mass measurement, the GFFs were equilibrated for 24 h at a constant temperature between 20 and 23°C and relative humidity between 35 and 45%. Each filter was weighed at least three times before and after sampling following the 24 h equilibration period. The mean net mass for each filter was obtained by subtracting the pre-deployment weight from the average of the post-sampling readings. The quartz PM_{10} sample filters were analyzed 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 pyrolyzed carbon fraction analyzed when reflected laser light attainsits original intensity after oxidation using a DRI Model 2001 Thermal and Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA). The protocol heats a 0.526 cm^2 punch aliquot of a QM/A stepwise at temperatures of 120°C (OC1), 250°C (OC2), 450°C (OC3) and 550°C (OC4) in a non-oxidizing helium atmosphere and 550°C (EC1), 700°C (EC2) and 800°C (EC3) in an oxidizing atmosphere of 2% oxygen in a balance of helium. For this study, OC and EC are defined as the sum of OC fractions (OC1+OC2+OC3+OC4) and EC fractions (EC1+EC2+EC3+OP), respectively, based on the Interagency Monitoring to Protect Visual Environments Total Organic Carbon protocol (IMPROVE TOC) (Chow *et al*., 1993, 2001; Fung *et al*., 2002).

RESULTS AND DISCUSSION

Like other capital cities in Southeast Asia, Bangkok experiences a tropical wet and dry climate under the Köppen climate classification. The rainy season starts with the arrival of the southwest monsoon around mid May, with an average precipitation of 344.2 mm in September and lasts until October (Table 1). The dry season begins when the dry and cool northeast monsoon takes over until February. Average and standard deviation values of OC and EC fractions in PM_{10} collected at eight PCD air quality observatory sites in Bangkok are illustrated in Table 2. Generally, the average concentrations for OC and EC were 10.0±2.70 and 10.1±4.28 μg m^{-3}, respectively. The ΣOC and ΣEC collected at eight PCD sites were 71.4±19.3 μg m^{-3} and 74.9±32.7 μg m^{-3}, correspondingly. It is interesting to note that both OC and EC demonstrated the highest average values at DHA. For instance, OC concentrations ranged from 10.4-20.1 μg m^{-3} with an average value of 14.2±2.71 μg m^{-3}, contributing on average, 17.7±12.5% to ΣOC. On the contrary, EC concentrations

Table 1: Climate data for Bangkok (1961-1990)

	Months												
Parameters	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Year
Record high (°C)[a]	35.7	36.6	37.8	40.0	39.5	37.7	37.8	37.0	36.0	35.3	35.1	35.2	40.0
Average high (°C)[a]	32.0	32.7	33.7	34.9	34.0	33.1	32.7	32.5	32.3	32.0	31.6	31.3	32.7
Daily mean (°C)[a]	25.9	27.4	28.7	29.7	29.2	28.7	28.3	28.1	27.8	27.6	26.9	25.6	27.8
Average low (°C)[a]	21.0	23.3	24.9	26.1	25.6	25.4	25.0	24.9	24.6	24.3	23.1	20.8	24.1
Record low (°C)[a]	11.5	14.9	15.7	19.9	21.1	21.7	22.2	21.2	21.7	18.3	14.2	10.5	10.5
Rainfall (mm)[a]	9.1	29.9	28.6	64.7	220.4	149.3	154.5	196.7	344.2	241.6	48.1	9.7	1497.0
Avg. rainy days (≥1 mm)[a]	1.0	3.0	3.0	6.0	16.0	16.0	18.0	20.0	21.0	17.0	6.0	1.0	128.0
Mean monthly sunshine (h)[b]	272.8	251.4	269.7	258.0	217.0	177.0	170.5	161.2	156.0	198.4	234.0	263.5	2630.0

[a]Thai Meteorological Department, [b]Hong Kong observatory (daily mean, sunshine) (http://www.hko.gov.hk/wxinfo/climat/world/eng/asia/se_asia/bangkok_e.htm)

Table 2: Statistical description of OC (OC1+OC2+OC3+OC4) and EC (EC1+EC2+EC3) in PM10 collected at eight PCD air quality observatory sites (μg m^{-3})

	OC1		OC2		OC3		OC4		OC	
Parameters	Aver*	SD**	Aver	SD	Aver	SD	Aver	SD	Aver	SD
BHS	0.005	0.005	1.896	0.367	4.673	1.277	2.296	0.723	8.869	2.281
KHA	0.006	0.005	1.845	0.336	4.381	1.139	2.287	0.737	8.519	2.106
NHS	0.009	0.016	1.935	0.277	4.741	1.235	2.481	0.537	9.166	1.900
WHS	0.007	0.008	1.848	0.440	4.803	1.680	2.299	0.703	8.957	2.756
EGAT	0.007	0.011	1.974	0.319	4.393	1.111	2.280	0.729	8.654	2.091
HOS	0.005	0.006	2.310	1.185	6.609	3.291	2.796	0.912	11.719	4.878
DHA	0.013	0.013	2.714	1.030	8.127	1.751	3.364	0.500	14.218	2.705
CPS	0.008	0.006	2.186	0.350	5.284	1.888	2.730	0.871	10.208	2.888
Aver	0.008	0.009	2.088	0.538	5.376	1.672	2.567	0.714	10.039	2.701
	EC1		EC2		EC3		EC		OC/EC	
Parameters	Aver	SD	Aver	SD	Aver	SD	Aver	SD	Aver	SD
BHS	5.614	1.505	0.472	0.230	0.068	0.203	6.154	1.522	1.441	1.499
KHA	6.091	1.677	0.321	0.149	0.021	0.036	6.433	1.782	1.324	1.182
NHS	8.382	2.318	0.325	0.158	0.009	0.012	8.715	2.352	1.052	0.808
WHS	5.882	2.348	0.279	0.163	0.02	0.026	6.181	2.447	1.449	1.126
EGAT	8.6	3.133	0.217	0.102	0	0.001	8.817	3.165	0.982	0.661
HOS	9.442	7.905	0.339	0.182	0.025	0.059	9.805	8.118	1.195	0.601
DHA	19.321	10.935	0.315	0.142	0.01	0.018	19.646	11.002	0.724	0.246
CPS	14.977	3.817	0.294	0.102	0.011	0.012	15.281	3.818	0.668	0.756
Aver	9.788	4.205	0.32	0.154	0.021	0.046	10.129	4.276	0.991	0.632

*Aver: Average, **SD: Standard deviation

varied from 6.18-19.6 μg m^{-3} with an average of 10.1±4.28 μg m^{-3}, contributing on average, 24.2±32.2% to ΣOC (Fig. 1). To determine whether the maximum values of ΣOC and ΣEC measured at DHA, are indeed due to significant emission source strengths or merely caused by coincidental effects of emissions, atmospheric transportation and chemical degradation, the statistical analysis was also performed.

By separating data set into eight groups (i.e., group-1: BHS (n = 11), group-2: KHA (n = 11), group-3: NHS (n = 11), group-4: WHS (n = 11), group-5: EGAT (n = 11), group-6: HOS (n = 10), group-7: DHA (n = 10) and group-8: CPS (n = 10)), significant levels of increase detected at DHA in both OC [$F(7, 77) = 5.17$, $p<0.05$] and EC [$F(7, 77) = 9.00$, $p<0.05$] were observed by using one way independent ANOVA. This finding is indeed consistent with a previous study revealing the comparatively high values of PAHs and Mutagenic Index (MI) detected at DHA (Pongpiachan *et al*., 2013b), suggesting that the single dominant source might have played an important role in governing pollutants in this area. According to the report by Office of Transport and Traffic Policy and Planning (OTP), Ministry of Transport, the five-year average (i.e., 2008-2012), traffic flow observed at Rama IX road (Zone 4E) was 14.0±1.3 km h^{-1}, while those of Ladprao road (Zone 8E), Petchburi road (Zone 5E), Prachachuen road (Zone 3N) and Phetkasem road (Zone 11W) were 13.8±1.8, 18.0±1.8, 19.2±1.5 and 20.4±6.8 km h^{-1}, respectively. The relatively low traffic flow indicates considerably high traffic congestion particularly at Rama IX road. Since DHA is located adjacent to Rama IX road, it seems rationale to interpret the comparatively high carbonaceous fractions measured at DHA as a contribution of vehicular

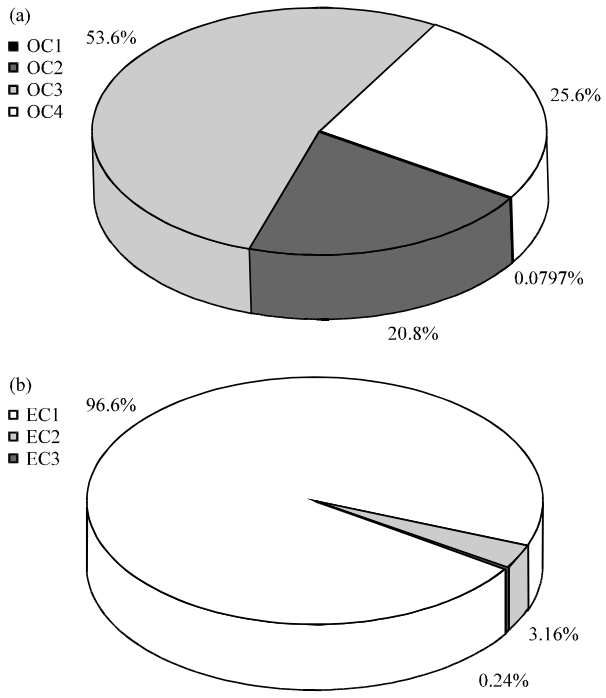

Fig. 1(a-b): Average values of percentage contributions of carbonaceous compositions (a) OC1, OC2, OC3, OC4 and (b) EC1, EC2 and EC3 measured at CPS, DHA, EGAT, HOS, KHA, NHS, WHS and BHS from February to December 2007

exhausts. This interpretation is further supported by an earlier study which reported the percentage of traffic contribution in Bangkok with a value of 86% (Pongpiachan, 2013a). Overall, these statistical results reveal the significant increase of carbonaceous compositions at the DHA. It is worth mentioning that confounding factors such as "re-suspension", "fluctuation of mixing layer depth", "variation of Long Range Atmospheric Transportation (LRAT)", "change of source fingerprint" and "chemical degradation" can have a huge impact on the variation of both OC and EC concentrations. Therefore, the data analysis must be performed with great caution. As illustrated in Table 3, the average OC concentration present in the samples taken in Bangkok (10.0±2.70 μg m^{-3}) is not only the highest on record but more than three times greater than the average values of whitbourne, UK (3.2±1.5 μg m^{-3}), Houtem, Belgium (3.2 μg m^{-3}) and Kosan, Korea (3.3±0.6 μg m^{-3}) while average EC concentrations were the highest in Xi'an, China (22.7±12.3 μg m^{-3}), followed by Shenzhen, China (10.4±6.8 μg m^{-3}) and Bangkok, Thailand (10.13±4.28 μg m^{-3}). The average EC concentrations for Bangkok were within the same range as those of Sihwa area, South Korea and Shenzhen, China.

Interpretation of OC/EC ratios: OC/EC ratios have been widely used to identify emission sources and investigate the photo-degradation mechanisms of carbonaceous species in particles. In this study, OC/EC ratios ranged from 0.43-2.28 with an average of 0.99±0.63. As illustrated in Table 2, OC/EC ratios in various studies reported from different locations worldwide, with Hong Kong, China (0.6±0.3) at the lowest end of the spectrum to Kosan, Korea (14.4±3.5) at the highest end. In general, the average OC/EC ratio in Bangkok is very similar to that of Chongju,

Table 3: Average OC and EC concentrations and the OC/EC ratios collected at various urban atmospheric environments

Sampling site	Area type	Period	OC (μg m^{-3})	EC (μg m^{-3})	OC/EC
Bangkok, Thailand[a]	Urban (Average values of 8 PCD sites)	February-December, 2007	10.04±2.70	10.13±4.28	0.991±0.632
Guangzhou, China[b]	Urban	August, 2006-August, 2007	7.1±3.3	4.0±2.5	1.8±1.4
Zhaoqing, China[b]	Semi-rural	August, 2006-August, 2007	8.2±5.0	3.9±2.0	2.1±1.7
Hong Kong, Polytechnic University, China[b]	Roadside	August, 2006-August, 2007	7.9±3.8	13.5±3.5	0.6±0.3
Hong Kong, Hok Tsui, China[b]	Rural	August, 2006-August, 2007	4.1±3.0	1.8±1.2	2.3±2.3
Whitbourne, UK[c]	Rural	November-December, 2003	3.2±1.5	1.1±0.5	2.9±1.9
Morogoro, Tanzania[d]	Rural	March-April, 2006	4.5±0.9	0.52±0.16	8.7±3.2
Zurich-Wiedikon, Switzerland[e]	City center	July/August, 1998 and January/February, 1999	9.0	7.7	1.2
Zurich-Kaserne, Switzerland[e]	City center	April, 1998-March, 1999	4.9	2.0	2.5
Basel, Switzerland[e]	Suburban	April, 1998-March, 1999	4.7	1.8	2.6
Payerne, Switzerland[e]	Rural	May/June and September/October, 1999	3.4	1.3	2.6
Bern, Switzerland[e]	City center	April, 1998-March, 1999	8.9	5.6	1.6
Chaumont, Switzerland[e]	Rural	April, 1998-March, 1999	1.7	0.6	2.8
Sihwa area, South Korea[f]	Urban-industrial	February, 1998-February, 1999	9.8±6.3	1.8±1.6	5.4±6.0
Houtem, Belgium[g]	Rural-background	September, 2006-September, 2007	3.2	0.5	6.8
Zelzate, Belgium[g]	Industrial site	September, 2006-September, 2007	4.4	1.3	3.4
Mechelen, Belgium[g]	Suburban-background	September, 2006-September, 2007	5.1	1.3	3.9
Borgerhout, Belgium[g]	Urban	September, 2006-September, 2007	5.3	2.0	2.6
Aarschot, Belgium[g]	Rural	September, 2006-September, 2007	4.1	1.0	4.2
Hasselt, Belgium[g]	Suburban	September, 2006-September, 2007	4.3	1.2	3.6
Thessaloniki, Greece[h]	Urban-industrial	December, 2006-March, 2007	8.7	2.9	3.0
Thessaloniki, Greece[h]	Urban-traffic	December, 2006-March, 2007	8.1	1.8	4.4
Cheju, Korea[i]	Urban-traffic	June-August, 1994	3.7	0.3	12.3
Kosan, Korea[j]	Rural-background	July-August, 1994	4.6±0.5	0.4±0.06	12.1±2.3
Seoul, South Korea[k]	Urban-background	June, 1999	10.3±3.2	8.4±3.0	1.2±0.6
Sihwa area, South Korea[k]	Urban-industrial	February, 1998-February, 1999	9.8±6.3	1.8±1.6	5.4±6.0
Hong Kong, China[l]	Urban-traffic	January-February, 2002	10.5±4.0	5.1±2.7	2.1±1.3
Guangzhou, China[l]	Urban-background	January-February, 2002	29.4±22.2	10.4±6.8	2.8±2.8
Shenzhen, China[l]	Urban-background	January-February, 2002	16.4±5.3	7.3±2.0	2.2±1.0
Thessaloniki, Greece[m]	Urban-industrial	June-September, 2007	6.4	2.9	2.2
Thessaloniki, Greece[m]	Urban-traffic	June-September, 2007	7.7	2.6	2.9
Kosan, Korea[n]	Rural	January, 1997	3.3±0.6	0.2±0.04	14.4±3.5
Chongju, South Korea[o]	Urban-industrial	October, 1995-August, 1996	5.0±2.4	4.4±2.7	1.1±0.9
Beijing, China[p]	Downtown	July-September, 1999	21.5	8.7	2.5
Zhuhai[q]	Urban-background	January-February, 2002	14.5±4.6	6.0±1.8	2.4±1.1
Xi'an, China[r]	Urban-background	September-October, 2003	43.2±27.1	15.0±10.7	2.9±2.7
Xi'an, China[r]	Urban-background	November, 2003-February, 2004	93.0±58.4	22.7±12.3	4.1±3.4
Tongliao, China[s]	Rural-background	March-May, 2005	15.7	3.3	4.8

[a]This study, [b]Huang *et al*. (2012), [c]Pongpiachan, 2006, [d]Mkoma *et al*. (2010), [e]Hueglin *et al*. (2005), [f]Park *et al*. (2001), [g]Vercauteren *et al*. (2011), [h]Terzi *et al*. (2010), [i]Kim *et al*. (1999), [j]Kim *et al*. (2000), [k]Park *et al*. (2005), [l]Cao *et al*. (2003), [m]Terzi *et al*. (2010), [n]Kim *et al*. (2000), [o]Lee and Kang (2001), [p]He *et al*. (2001), [q]Cao *et al*. (2003), [r]Cao *et al*. (2005), [s]Li *et al*. (2006)

South Korea (1.1±0.9) but lower than that of Morogoro, Tanzania (8.7±3.2) and Cheju, South Korea (12.3). During the past two decades, several studies have attempted to determine the significance of OC/EC ratios collected from aerosol observatory sites around the world. For instance, Sawant *et al.* (2004) suggested that the high OC/EC ratio in $PM_{2.5}$ in Mira Loma is due to high OC mass concentration rather than low EC mass concentrations. Conversely, Kim *et al.* (2000) attributed the high OC concentration in inland areas such as Rubidoux to the formation of secondary OC while the high OC level in downtown Los Angeles was more likely due to primary OC. The OC/EC ratios obtained from the Bangkok samples at all PCD observatory sites (0.99±0.6) ranged from those of heavy-duty diesel vehicles (0.8) to light-duty gasoline vehicles (2.2) but much lower than those of natural gas home appliances (12.7), paved road dust (13.1) and forest fire (14.5) (Hildemann *et al.*, 1991; Watson *et al.*, 1994; 2001). It is worth mentioning that this result is not consistent with a study conducted by Na *et al.*, (2004) which showed a weak correlation coefficient (R^2 = 0.50) between organic and elemental carbon and average OC/EC ratio (5.2) for California aerosols ranging from residential wood combustion (4.15) to forest fire (14.51). The reason for this difference could possibly be attributed to the existence of more complex source strengths in the California atmospheric environment than those in Bangkok. It is also interesting to note that relatively high OC/EC ratios have also been attributed to certain factors in particular. Firstly, a high OC/EC ratio is often associated with the existence of secondary OC derived from photochemical reactions. However, it is difficult to conclude the presence of secondary OC from absolute values of OC/EC alone since the ratios can be affected by various other causes such as meteorological conditions or diurnal and seasonal fluctuations in emissions. Secondly, it is largely accepted that in a clean rural environment, emissions of EC are limited and thus the OC/EC ratio tends to be high (Satsangi *et al.*, 2012). Thirdly, agricultural waste burning and forest fires can cause a higher OC with a lower EC emission rate and thus produce higher OC/EC values (Pongpiachan *et al.*, 2009). Hence, it appears reasonable to ascribe relatively low OC/EC ratios observed in Bangkok as a result of (1) Less activities in photochemical reactions (i.e., less OC derived from photochemical reactions), (2) More polluted air from traffic exhausts (i.e., emissions of EC are high and thus the OC/EC ratio tends to be low) and (3) Non-biomass burning activities observed adjacent to the sampling sites.

In order to explain the relatively low OC/EC ratios in Bangkok, one may use the relationship between OC and EC atmospheric concentrations to qualitatively evaluate the source of carbonaceous particles (Turpin *et al.*, 1994). If major fractions of OC and EC are introduced into atmospheric environment by a single primary source (e.g., vehicle emissions), the correlation coefficients (R^2-values) between the OC and EC concentrations should be close to one because the relative rates of EC and OC emissions would be proportional to each other. In the present study, a considerably strong positive correlation coefficient (R^2 = 0.61, $p<0.001$) was found between organic and elemental carbon at all sites. As illustrated in Table 4, R^2-values from eight different PCD observatory sites, with NHS (R^2 = 0.42) at the minimum end of the spectrum to HOS (R^2 = 0.85) at the maximum, followed by DHA (R^2 = 0.84) and BHS (R^2 = 0.77). Since HOS and DHA are located in heavy traffic congested areas, the comparatively high R^2-values can be interpreted as a consequence of a single, dominant primary source. On the contrary, the relative low R^2-values detected at NHS (0.42) and EGAT (0.53) are more likely to be influenced by several sources in comparison to those of HOS and DHA.

Table 4: Slopes, intercepts and R-values obtained from linear regression between OC and EC

Parameters	Slope (m)	Intercept (b)	R^2
BHS	0.659	0.439	0.768
KHA	0.963	-1.554	0.680
NHS	0.897	0.809	0.418
WHS	0.901	-1.796	0.740
EGAT	1.200	-1.262	0.525
HOS	1.534	-8.170	0.849
DHA	3.720	-33.243	0.836
CPS	1.079	4.268	0.666
Aver	1.610	-6.085	0.610

Estimation of secondary organic carbon: Further estimation of secondary organic carbon in Bangkok PM_{10} was conducted by using the method reported by Na *et al.* (2004). The concept of this method is based on the assumption that samples having the lowest OC/EC ratios contain almost exclusively primary carbonaceous compounds (Castro *et al.*, 1999). For this study, OC/EC ratios of 0.51, (the average of the three lowest OC/EC ratios), was employed to estimate the Secondary Organic Carbon (SOC) content of Bangkok PM_{10}. Although carbonaceous samples with minimum OC/EC ratios are expected to contain exclusively primary OC, a small proportion of secondary OC may still exist. Therefore, the results of this OC/EC method provide a lower limit for SOC content. The concentration of Secondary Organic Carbon (SOC) can be estimated using the subsequent equation:

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

where, $(OC/EC)_{primary}$ is the average value of the three lowest OC/EC ratios and OC_{tot} is the total organic carbon.

During the study period, the OC and EC concentrations varied significantly from period to period, having lowest values of 5.26 μg m^{-3} (03/04/07: WHS) and 2.31 μg m^{-3} (03/04/07: WHS) and highest values of 23.1 μg m^{-3} (06/09/07: HOS) and 46.6 μg m^{-3} (11/12/07: DHA) for OC and EC, respectively. By using Eq. 1 to calculate OC_{sec} concentrations in Bangkok aerosols, the percentage contributions of OC_{sec} to OC_{tot} varied from 5.5-77.6% with the average value of 52.2±17.5%. This average value is almost 1.3 times higher than observed in Kaohsiung (40.0%, Lin and Tai, 2001) and approximately three times higher than found in Birmingham, UK (17%, Castro *et al.*, 1999) but lower than measured in Claremont, US $PM_{2.5}$ (65%, Na *et al.*, 2004). This indicates the substantial contribution of OC_{sec} to OC_{tot} and in turn, to total PM_{10} mass in Bangkok.

Seasonal effects on variation of carbonaceous contents: The fate and behavior of carbonaceous compositions in the atmospheric environment has attracted substantial scientific and political interest, arising from concern over human exposure to these chemicals and their discovery in remote area far from the source regions. The atmosphere is clearly an important media in conveying carbonaceous compositions to the oceans and rivers then subsequently to sediments. After their emission to the atmosphere, carbonaceous compositions are subject to a variety of atmospheric processes which can be summarized as follows: (1) Physical removal by dry and wet deposition, (2) Atmospheric transport and dispersion, (3) Atmospheric degradation and transformation by chemical and photochemical reactions and (4) Shifts in the phases equilibrium

between the gaseous and particulate phase due to changes in the ambient temperature. Several factors which govern the fate and behavior of carbonaceous aerosols in atmosphere, are the vapor pressure, water solubility and chemical reactivity.

Atmospheric deposition is an important mechanism controlling the fate of carbonaceous compositions and their transfer from the atmosphere to natural surfaces. Dry deposition is the transport of gaseous and particulate species from the atmosphere onto surfaces in the absence of precipitation. Dry deposition is much slower than wet deposition but it acts continuously rather than episodically. The factors that govern the dry deposition of a gaseous species or particle are the level of atmospheric turbulence, the chemical properties of the deposition species and the nature of the surface itself. For gases, solubility and chemical reactivity may affect uptake at the surface. For particles, size, density and shape may determine whether capture by the surface occurs. The surface itself is also a factor in dry deposition. Wet deposition refers to the natural processes by which material is scavenged by atmospheric hydrometeors (cloud, fog drops, rain and snow) and is consequently delivered to the Earth's surface. The processes occur during atmospheric wet deposition can be summarized as follows: (1) Precipitation scavenging, that is, the removal of chemical species by a raining cloud, (2) Cloud interception, the impaction of cloud droplets on the terrain usually at the high mountains, (3) Fog deposition, that is, the removal of chemical compounds by setting for droplets and (4) Snow deposition, removal of chemical compounds during a snowstorm. Like dry deposition, wet deposition is one of the most significant processes in removing carbonaceous compositions from atmospheric environment.

As previously mentioned, Bangkok has only dry and rainy seasons under the Köppen climate classification. Therefore, it appears reasonable to compare the atmospheric contents of OC and EC measured in both periods. In order to investigate the seasonal effects on particulate OC and EC, the two-sample-t-test of carbonaceous compositions between two seasons in 2007 was conducted as illustrated in Table 5. The ratios of carbonaceous contents collected in dry and rainy seasons (i.e., D/R ratios) for all individual carbonaceous compositions were generally higher than one except for OC1 and EC2. In this sense, it can be hypothesized that the "wash out" phenomenon might have played an important role in reducing carbonaceous contents in Bangkok's atmosphere. In order to test this hypothesis, the average carbonaceous concentrations of both periods were subjected to an independent t-test to see if there has been any evidence of a significant change in individual composition between the two seasons (Table 4). In contrast to the results of D/R ratios

Table 5: Two-sample-t-test of carbonaceous compositions between rainy and dry seasons in 2007

	Rainy season (May-October)		Dry season (November-April)		D/R ratio			
Parameters	Aver	SD	Aver	SD	Aver	SD	t-value	p<0.05
OC1	0.008	0.010	0.007	0.009	0.976	0.964	0.932	NS
OC2	1.969	0.598	2.218	0.707	1.126	1.182	0.090	NS
OC3	4.954	2.033	5.819	2.097	1.175	1.031	0.060	NS
OC4	2.387	0.735	2.768	0.791	1.160	1.077	0.026	NS
OC	9.317	3.082	10.812	3.332	1.160	1.081	0.037	NS
EC1	9.305	5.716	10.027	7.826	1.078	1.369	0.638	NS
EC2	0.332	0.131	0.305	0.206	0.920	1.565	0.497	NS
EC3	0.014	0.031	0.029	0.112	2.007	3.564	0.449	NS
EC	9.651	5.755	10.361	7.858	1.074	1.365	0.646	NS

NS: Not significant at the confidence level of 95%, D/R ratio: Dry/rainy season ratio

which seem to have values higher than one, Table 4 shows the "non-significant" level ($p<0.05$) of "rainy season decline" for each individual carbonaceous composition in eight PCD air quality observatory sites in Bangkok. It seems likely that primary carbonaceous emissions from road traffic exceed the washout phenomenon and thus the effects of "wet deposition" by precipitation were not observed in this area.

Multiple Linear Regression Analysis (MLRA): The variation of gaseous PAHs depends mainly on meteorological variables such as ambient temperature, wind speed and wind direction (Dachs *et al*., 2002). For particulate PAHs, the chemical compositions, namely, the content of OC and EC could be responsible for the variations. In order to investigate the influence of carbonaceous fractions and meteorological variables, PAH aerosol concentrations have been fitted by multiple linear regressions to:

$$C_{aerosol} = a+bT+cU_{10}+dsin(WD)+ecos(WD)+fC_{OC}+gC_{EC} \quad (2)$$

where, $C_{aerosol}$, T, U_{10}, WD, C_{OC} and C_{EC} stand for PAH aerosol concentration, ambient temperature, wind speed, wind direction, OC and EC contents, respectively. A similar approach to that of Dachs *et al*. (2002) was taken by modified Eq. 2 with four trace gaseous species namely CO, NO_x, SO_2 and O_3:

$$C_{OC} = a+bT+cU_{10}+dsin(WD)+ecos(WD)+fC_{CO}+gC_{NO_x}+hSO_2+iO_3 \quad (3)$$

$$C_{EC} = a+bT+cU_{10}+dsin(WD)+ecos(WD)+fC_{CO}+gC_{NO_x}+hSO_2+iO_3 \quad (4)$$

Multiple linear regressions can establish the relative predictive importance of the independent variables, namely, T, U_{10}, WD and trace gaseous concentrations on the dependent variables (i.e., C_{OC} and C_{EC}). Calculations were made using software SPSS 13.0 for Microsoft Windows with the 'stepwise' MLRA method. Table 6 summarized the regression coefficients obtained from Eq. 3 and 4. Regression coefficient is the average amount the dependent increases when the independent increases one unit and other independents are held constant. Thus the higher the coefficients, the more influence there is on carbonaceous aerosol concentrations. On the other hand, β is the average amount the dependent increases when the independent increases on standard deviation and other independent variables are held constant. Therefore, the β weight reflects the unique contribution of each independent variable to OC/EC concentrations. As can be seen in Table 6, OC compositions had significant positive regression weights with the value of 0.396 ($p<0.015$), indicating air samples with higher NO_x concentration on these scales were expected to have higher OC contents in PM_{10} samples. Similarly, EC compositions had shown significant positive β weights of 0.390 ($p<0.009$) and 0.387 ($p<0.007$) for NO_x and SO_2, respectively, highlighting the influence of vehicle exhaust emissions on the enhancement of elemental carbon in the Bangkok atmosphere.

Principal Component Analysis (PCA): Principal Component Analysis (PCA) is employed as the multivariate analytical tool to reduce a set of original variables (i.e., measured carbonaceous content in PM_{10} samples and meteorological parameters) and to extract a small number of latent factors (PCs) to analyze relationships among the observed variables. Data submitted for analysis

Table 6: Multiple linear regressions of carbonaceous particles and meteorological parameters

	Unstandardized coefficients		Standardized coefficient		
Dependent variable	B	Std. error	Beta	t	Significance
OC					
Constant	12.134	4.777		2.540	0.014
T	-0.206	0.153	-0.131	-1.342	0.184
WS	-0.742	0.546	-0.133	-1.358	0.179
Sin (WD)	0.037	0.571	0.007	0.065	0.948
Cos (WD)	-0.035	0.792	-0.005	-0.044	0.965
CO	1.796	1.139	0.260	1.577	0.120
NO_x	0.119	0.047	0.396	2.501	0.015
SO_2	0.009	0.106	0.007	0.083	0.934
O_3	0.037	0.036	0.098	1.015	0.314
EC					
Constant					
T	0.682	8.818		0.077	0.939
WS	0.046	0.283	0.014	0.162	0.871
Sin (WD)	0.516	1.008	0.044	0.512	0.611
Cos (WD)	1.452	1.053	0.126	1.378	0.173
CO	0.786	1.461	0.053	0.538	0.592
NO_x	5.625	2.102	0.390	2.676	0.009
SO_2	0.242	0.088	0.387	2.763	0.007
O_3	0.055	0.196	0.022	0.280	0.780

Table 7: Principal Components (PC) pattern for Varimax rotated components applied to carbonaceous compositions, trace gaseous species and meteorological data set from the eight PCD air quality observatory sites

	Principal component (PC)		
Parameters	1	2	3
OC	0.852	0.148	-0.126
EC	0.870	-0.181	-0.093
NO_x	0.869	0.197	-0.213
CO	0.888	-0.041	0.002
O_3	-0.124	0.794	-0.217
SO_2	0.136	-0.070	0.827
WS	-0.476	-0.179	-0.160
WD	-0.239	-0.178	0.477
RH	-0.416	-0.673	0.133
T	-0.457	0.434	0.404
P	0.094	0.751	0.005
Total of variance (%)	35.4	18.0	10.0

was arranged in a matrix, where each column corresponds to one parameter component and each row represents the number of samples. Data matrixes were evaluated through PCA, allowing the summarized data to be further analyzed and plotted. Table 7 displays the principal component patterns for Varimax rotated components of the carbonaceous data set at all observatory sites, coupled with anion species, trace gaseous species and meteorological parameters. In order to further interpret potential carbonaceous sources, a PCA model of all sampling sites with three significant PCs representing 35.4% (PC1), 18.0% (PC2) and 10.0% (PC3) of the variance, thus accounting for

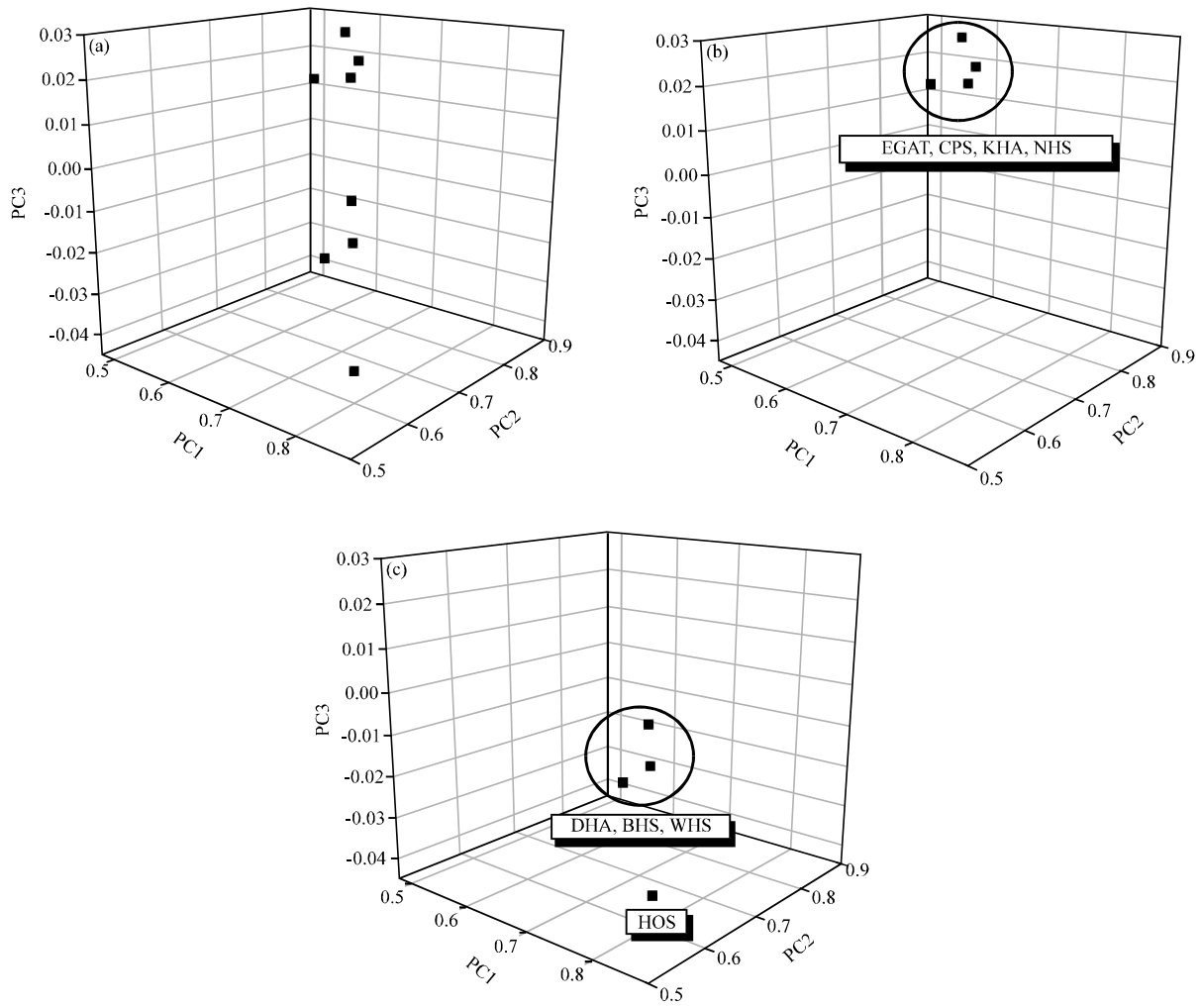

Fig. 2(a-c): Three-dimensional plots of PC1, PC2 and PC3 obtained from carbonaceous compositions (OC1, OC2, OC3, OC4, EC1, EC2 and EC3) measured at CPS, DHA, EGAT, HOS, KHA, NHS, WHS and BHS from February to December 2007

63.4% of the total variation in the data, was calculated. The first component (PC1) shows high loading on OC (R = 0.85), EC (R = 0.87), NO_x (R = 0.87) and CO (R = 0.89). Since NO_x and CO can be used as indicators of the contribution of imperfect combustion from vehicle exhausts, the considerably strong positive correlation between these two trace gaseous species and carbonaceous components indicate the major contribution of traffic emissions to OC and EC in PC1. Further attempts on data classification were made by using PCA with data matrix of seven carbonaceous fractions and eight PCD observatory sites. The principal component patterns for Varimax rotated components were composed of three components, namely PC1 (97.4%), PC2 (2.54%) and PC3 (0.063%) which accounted for 99.99% of the total variance. As illustrated in Fig. 2, three-dimensional plots of PC1, PC2 and PC3 obtained from carbonaceous compositions (OC1, OC2, OC3, OC4, EC1, EC2 and EC3) measured at CPS, DHA, EGAT, HOS, KHA, NHS, WHS and BHS from February to December 2007, were displayed. The clearest features in all

categories (Fig. 2) are: (1) 3D plots of EGAT, CPS, KHA and NHS, (2) There are very clear different sources in air samples of DHA, BHS and WHS; (3) 3D plots of HOS is highly deviated from both groups.

Occupational exposure to carbonaceous PM_{10} of outdoor workers: In this study, the ILPE model was used to investigate the potential health risk related with the occupational exposure to carbonaceous PM_{10} of out-door workers adjacent to the eight PCD observatory sites. The model is described in Eq. 5:

$$ILPE = C \times IR \times t \times EF \times ED \quad (5)$$

Where:

ILPE = Incremental lifetime particulate matter exposure (g)
C = $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$)
IR = Inhalation rate ($m^3\ h^{-1}$)
t = Daily exposure time span (6 h day^{-1}, for two shifts)
EF = Exposure frequency (250 day $year^{-1}$[a], upper-bound value)
ED = Exposure duration (25 years[a], upper-bound value)
[a]Adapted from Human Health Evaluation Manual (USEPA, 1991)

According to the methods for derivation of inhalation dosimetry (USEPA, 1994), the inhalation rate of male and female out-door workers were estimated as 0.89 and 0.49 $m^3\ h^{-1}$, respectively. The ILPE model was adapted from the probabilistic Incremental Lifetime Cancer Risk (ILCR) model which was used to assess traffic policemen exposure to PAHs during their work time in China (Hu *et al*., 2007). The estimated ILPE levels in outdoor workers are summarized in Table 8. The estimated ILPE of carbonaceous compositions were constantly highest at DHA in both genders with the average values of 1,130±457 and 622±252 mg for TC accumulated in male and female workers

Table 8: Incremental lifetime exposure of OC (OC1+OC2+OC3+OC4) and EC (EC1+EC2+EC3) in PM_{10} collected at eight PCD air quality observatory sites

	Male						Female					
	TC (mg)		OC (mg)		EC (mg)		TC (mg)		OC (mg)		EC (mg)	
Parameters	Aver	SD	Aver	SD	Aver	SD	Aver	SD	Aver	SD	Aver	SD
BHS	501	127	296	76	205	51	276	70	163	42	113	28
KHA	499	130	284	70	215	59	275	71	157	39	118	33
NHS	597	142	306	63	291	79	329	78	168	35	160	43
WHS	505	174	299	92	206	82	278	96	165	51	114	45
EGAT	583	175	289	70	294	106	321	97	159	38	162	58
HOS	718	434	391	163	327	271	396	239	215	90	180	149
DHA	1130	457	475	90	656	367	622	252	261	50	361	202
CPS	851	224	341	96	510	127	468	123	188	53	281	70
Average	673	233	335	90	338	143	371	128	184	50	186	79

Department of health 2005 (average body weight). Male: 58.25±9.76 kg, Female: 54.95±10.48 kg. Life Expectancy: National Statistical Office Thailand (1996) (survey of population change 1995-1996). Male: 69.9 year, Female: 74.9 year. Inhalation Rate, Male: 0.89 $m^3\ h^{-1}$ (USEPA, 1994), Female: 0.49 $m^3\ h^{-1}$

over exposure duration of 25 years, respectively. The second highest ILPE-TC in PM_{10} samples were detected at CPS in both genders with the average values of 851±224 and 468±123 mg for male and female, respectively. Interestingly, HOS shows the third highest ILPE-TC values with the average values of 718±434 and 396±239 mg for male and female, respectively. These results suggest that those out-door workers in traffic congested areas are potentially exposed to a wide variety of carbonaceous compositions, raising concerns over long-term adverse respiratory effects. It is also important to note that the average ILPE-TC values of "traffic emissions" group (i.e., CPS, DHA, EGAT, HOS) are 1.56 times higher than those of "urban residential background" group (i.e., KHA, NHS, WHS, BHS) for both genders.

CONCLUSION

Irrespective of sampling locations, the comparatively low OC/EC ratios highlight significant contribution from traffic emissions. Significant levels of increase detected at DHA in carbonaceous compositions indicating that the vehicular exhausts might have played a crucial role in controlling carbonaceous compositions in heavy traffic congestion area. No significant seasonal effects on either OC or EC were observed which suggests that neither "dry deposition" nor "wet deposition" influences on both spatial and temporal distribution of carbonaceous aerosols. The relatively high β weight obtained from MLRA reflects the unique positive correlations between NO_x and carbonaceous fractions indicating that imperfect combustions of automobile engine could have been a major source of OC/EC fractions. This finding is consistent with PCA results which accounting for 63.4% of the total variation in three principal components. Since the first component (PC1) shows high loading on OC, EC, No_x and CO, it appears rationale to conclude this finding as a consequence of an overwhelming influence of transportation sector on air quality level in Bangkok.

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