RESEARCH ARTICLE

Estimation of Gas-Particle Partitioning Coefficients (K_p) of Carcinogenic Polycyclic Aromatic Hydrocarbons in Carbonaceous Aerosols Collected at Chiang-Mai, Bangkok and Hat-Yai, Thailand

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Abstract

To assess environmental contamination with carcinogens, carbonaceous compounds, water-soluble ionic species and trace gaseous species were identified and quantified every three hours for three days at three different atmospheric layers at the heart of Chiang-Mai, Bangkok and Hat-Yai from December 2006 to February 2007. A DRI Model 2001 Thermal/Optical Carbon Analyzer with the IMPROVE thermal/optical reflectance (TOR) protocol was used to quantify the organic carbon (OC) and elemental carbon (EC) contents in PM_{10}. Diurnal and vertical variability was also carefully investigated. In general, OC and EC mass concentration showed the highest values at the monitoring period of 21.00-00.00 as consequences of human activities at night bazaar coupled with reduction of mixing layer, decreased wind speed and termination of photolysis at nighttime. Morning peaks of carbonaceous compounds were observed during the sampling period of 06:00-09:00, emphasizing the main contribution of traffic emission in the three cities. The estimation of incremental lifetime particulate matter exposure (*ILPE*) raises concern of high risk of carbonaceous accumulation over workers and residents living close to the observatory sites. The average values of incremental lifetime particulate matter exposure (*ILPE*) of total carbon at Baiyoke Suit Hotel and Baiyoke Sky Hotel are approximately ten times higher than those air samples collected at Prince of Songkla University Hat-Yai campus corpse incinerator and fish-can manufacturing factory but only slightly higher than those of rice straw burning in Songkla province. This indicates a high risk of developing lung cancer and other respiratory diseases across workers and residents living in high buildings located in Pratunam area. Using knowledge of carbonaceous fractions in PM_{10}, one can estimate the gas-particle partitioning of polycyclic aromatic hydrocarbons (PAHs). Dachs-Eisenreich model highlights the crucial role of adsorption in gas-particle partitioning of low molecular weight PAHs, whereas both absorption and adsorption tend to account for gas-particle partitioning of high molecular weight PAHs in urban residential zones of Thailand. Interestingly, the absorption mode alone plays a minor role in gas-particle partitioning of PAHs in Chiang-Mai, Bangkok and Hat-Yai.

Keywords: Carbonaceous aerosols - vertical distribution - gas-particle partitioning coefficient (K_p) PAHs - Thailand

Asian Pacific J Cancer Prev, **14 (4)**, 2461-2476

Introduction

Carbonaceous aerosols, generally recognized as organic carbon (OC) and elemental carbon (EC) particles, have been intensively studied in the past decades because these atmospheric particles offer a wide range of impacts on both health and climate system (Penner et al., 1996; O'Brien and Mitchell, 2003; Shih et al., 2008). Carbonaceous aerosol is detrimental to human health, according to recent study on effects of fine carbonaceous particles containing high and low unpaired electron spin densities on lungs of female mice (Repine et al., 2008). As a consequence of concern over their existence in the atmospheric environment, several studies were conducted to investigate its influences on gas-particle partitioning of carcinogenic and/or mutagenic substances such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and pesticides (Simcik et al., 1998; Kaupp and McLachlan, 1999; Dachs and Eisenrich, 2000; Schummer et al., 2010). These carbon-containing compounds are often referred to as persistent organic pollutants (POPs) that are persistent to photochemical degradation, biological decomposition, and chemical reactions (Jones and Voogt, 1999).

The character of carbonaceous aerosols, which are

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composed of organic carbon (OC) and elemental carbon (EC) particles, has been continuously investigated during the past three-decades (Ellis and Novakov, 1982; Lammel and Novakov, 1995; Park et al., 2001; Ho et al., 2002; Dan et al., 2004; Pathak et al., 2011). Several concerns over their adverse impact on both human health and the climate have been raised by scientists around the World reflecting their importance (Penner et al., 1996; O'Brien and Mitchell, 2003; Repine et al., 2008; Shih et al., 2008). Carbonaceous aerosols, particularly elemental carbon (EC), have been considered as factors affecting climate system due to its atmospheric radiative forcing capability (Badarinath and Latha, 2006; Panicker et al. 2010; Cheng and Wu, 2011; Das and Jayaraman, 2011). Apart from its impacts on climate system, the role of carbonaceous particles on gas-particle partitioning of persistent organic pollutants (POPs) such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), pesticides and dioxins, has been extensively evaluated in different countries during the past few years (Hayakawa et al., 2004; Tasdemir et al., 2004; Scheyer et al., 2008; Pongpiachan et al., 2009; Marcosa et al., 2010; Gaga and Ari, 2011; Yang et al., 2012; Pongpiachan, 2013a; 2013b). Since POPs are widely considered as carcinogenic and mutagenic compounds (Binelli and Provini, 2004; Abas and Mohamad, 2011; Guo et al., 2012), it is therefore crucial to understand the behavior of OC/EC compositions in atmospheric environment.

According to circumstances, there have been a growing number of studies focusing on the analysis of chemical compositions of carbonaceous aerosols collected at observatory sites around the world (Cao et al., 2004; Na et al., 2004; Han et al., 2008; Ram et al., 2008). 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. 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 (Fang et al., 1999; Latha and Badarinath, 2003; 2005; Pandey et al., 2006) while others have examined the empirical estimation of secondary organic carbon (SOC) formation (Seguel et al., 2009; Grivas et al., 2012; Kim et al., 2012; Wang et al., 2012). Only a few publications have presented in-depth, quantitative evidence regarding the behavior of carbonaceous aerosols in Thailand (Pongpiachan et al., 2009; Sahu et al., 2011; Li et al., 2012).

During the past decades, a comprehensive physicochemical investigation of the gas-particle partitioning has been conducted in order to predict the K_p of PAHs. By assuming that the gas-particle partitioning of PAHs was governed by *ab*sorption process, the K_p values can be estimated by using only the knowledge of octanol/air partition coefficient (K_{OA}) as written in Equation 1 (Finizio et al., 1997).

$$Kp=f_{OM}[(MW_{oct}\gamma_{oct})/(MW_{OM}\gamma_{OM}10^{9}\rho_{oct})]K_{oa} \quad \textbf{Equation 1}$$

Where ρ_{oct} (820 kg m^{-3} at 20°C) is the density of the octanol and γ are the activity coefficients of the PAHs in organic matter (OM) and octanol (oct), respectively. Assuming that *ab*soption is the dominant sorption process and $MW_{oct}/MW_{OM}=\gamma_{oct}/\gamma_{OM}=1$, Equation 1 can then be simplified to

$$LogK_p=LogK_{OA}+Logf_{OM}-11.91 \quad \textbf{Equation 2}$$

Fundamentally, Equation 2 was developed from the Pankow model where both K_p and P_L^o were used as descriptors for absorptive partitioning.

$$Kp=f_{OM}760RT/MW_{OM}P_L^o \quad \textbf{Equation 3}$$

where f_{OM} is the fraction of OM and ζ are the activity coefficients of PAHs in OM.

In cases when the dominant partitioning process is not *ab*sorption into the OM phase, then it seems reasonable to consider about a prominent role of elemental carbon (EC) *ad*sorption in the atmosphere. Several evidences strongly support the possibility that *ad*sorption onto the soot phase controlling the sorption mechanism of PAHs in the New Jersey atmosphere (Dachs and Eisenreich., 2000). Since EC is considered to be a surrogate for the soot phase, the overall sorption mechanisms that includes for both the *ab*sorption into OM phase and the *ad*sorption on soot phase is given by

$$K_p=[(f_{om}MW_{OCT}\zeta_{OCT})/(\rho_{oct}MW_{om}\zeta_{OM}10^{12})]K_{OA}+f_{ec}[(a_{EC}/10^{12}a_{AC})]K_{SA} \quad \textbf{Equation 4}$$

where f_{EC} is the fraction of elemental carbon in the aerosol, a_{AC} is the surface area of the activated carbon, a_{AE} is the specific surface area of the elemental carbon and K_{SA} is the soot-air partitioning coefficient. When the partitioning is occupied by both *ad*sorptive and *ab*sorptive processes, K_p can be accurately predicted by using the Dachs-Eisenreich model (Equation 4). Although many researchers highlight the role of *ad*sorption onto black carbon (BC) surfaces as a major sorption mechanism, only few middle MW PAH compounds were investigated. Therefore, little is known so far about the gas-particle partitioning processes of low and high MW PAH compounds as it can be explained by the lack of experimental K_{SA} values (Ribes et al. 2003). Furthermore, it is well known that both K_{SA} and K_{OA} can be significantly influenced by temperature which lead to a serious necessity to minimize any uncertainties caused by the temperature fluctuation.

Recently, Odabasi et al. (2006) determined octanol–air partition coefficients (K_{OA}) and super-cooled liquid vapor pressure (P_L) for 13 PAHs namely, acenaphthylene (Ac), (Ace), fluorene (Fl), phenanthrene (Ph), anthracene (An), fluoranthene (Fluo), pyrene (Pyr), benzo[*a*]anthracene (B[*a*]A), chrysene (Chry), benzo[*a*]pyrene (B[*a*]P), indeno[*1,2,3-cd*]pyrene (Ind), dibenz[*a,h*]anthracene (D[*a,h*]A), and benzo[*g,h,i*]perylene (B[*g,h,i*]P) as a function of temperature using the gas chromatographic retention time technique. The temperature dependence of K_{OA} can be obtained by

$$logK_{OA}=A+(B/T) \quad \textbf{Equation 5}$$

where *A* and *B* were calculated by Harner and Bidleman (1998a; 1998b). In this study, K_{SA} values for PAHs were estimated as a function of super-cooled liquid vapor pressure (P_L) and elemental carbon specific surface area (a_{EC}, m^2g^{-1}) as can be displayed in Equation 6 described by Odabasi et al. (2006):

$$logK_{SA}=-0.85logP_L+8.94-log(998/a_{EC}) \quad \textbf{Equation 6}$$

where the a_{EC} value of 62.7 m^2 g^{-1} was reported by Jonker and Koelmans (2002) and P_L can be obtained by using the intercept b_L, slope m_L and temperature T as can be expressed by Equation 7 (Odabasi et al., 2006).

$$logP_L(Pa)=m_L(T,K)^{-1}+b_L \quad \textbf{Equation 7}$$

In the present work, we reported the gas-particle partitioning coefficients (K_P) of PAHs estimated by using the Dachs-Eisenreich model with the measured OC/EC values of the small particles-PM_{10} (aerodynamic diameter <10 μm) from three different sampling altitudes at Chiang-Mai, Bangkok and Hat-Yai. The paper is aimed to quantify the relative contribution of carbonaceous aerosols, and to estimate the K_P values of PAHs in PM_{10} from three major cities in Thailand. The results obtained in this study allow assessing the impact of air quality and assist the policy maker to make a correct policy decisions and control strategies for occupational inhalation disease reduction.

Overall, the main purposes of this study are i) To analyze the OC/EC ratios and interpret the impacts of meteorological parameters to the fate of carbonaceous compositions in PM_{10}. ii) To calculate the incremental lifetime particulate matter exposure (*ILPE*) of carbonaceous aerosols at three different heights. iii) To estimate the K_P values of PAHs in PM_{10} collected from three major cities in Thailand.

Materials and Methods

Sampling sites

Bangkok: Bangkok is one of the most densely populated capital cities in Thailand with approximately 12 million people. Bangkok has 50 districts under the authority of the Bangkok Metropolitan, covering administrative area of 1,568.7 km^2, making it the 73rd largest city in the world. It is well known that Bangkok is experiencing gradually increased air quality problems associated with high traffic emissions, causing adverse effects on human health (Muttamara and Leong, 2002). Although the air quality levels in Bangkok are generally acceptable, the roadside measurement of toxic gaseous species are sometime observed to exceed the national air quality standard. As a consequence, a number of studies attempted to investigate the behavior of carcinogenic substances in airborne particles of Bangkok's atmosphere (Chetwittayachan et al., 2002; Boonyatumanond et al., 2007; Hoshiko et al., 2011). As there were only a few studies providing information of carbonaceous compositions in aerosols collected at urban cities of Thailand (Pongpiachan et al., 2009; Sahu et al., 2011; Li et al., 2012), it is difficult to conduct the evaluation of human health impact caused by exposure to carbonaceous aerosols for Thai citizens.

Two observatory stations, namely Baiyoke Suit Hotel (13°45'10.65" N 100°32'24.92" E) and Baiyoke Sky Hotel (13°45'15.69" N 100°32'29.73" E), were situated in the center of Bangkok Metropolitan encompassed by cloth markets and shopping areas. Three different observatory stations were cautiously chosen consistent with its height levels. Site-1 and site-2 were situated at Baiyoke Suit Hotel at 38 m and 158 m above ground level respectively, whilst site-3 was located at Baiyoke Sky Hotel at 328 m above the building basement. It is also worth to mention that Baiyoke Sky Hotel is the tallest building in Southeast Asia, and the fourth-tallest all-hotel structure in the world. Intensive monitoring campaigns were conducted at all observatory stations instantaneously from February 18th to 21st 2008 in cold period. PM_{10} samples were collected every three hours consecutively from 2100 h February 18th to 2100 h February 21st by using Graseby-Andersen High Volume Air Sampler PM_{10} TE-6001 with the flow rate of 1.132 m^3 min^{-1}. More detailed description of the air sampling method were provided in "Compendium Method IO-2.2. Sampling of Ambient Air for PM_{10} Using an Andersen Dichotomous Sampler".

Chiang-Mai: There have, however, been an increasing number of articles to appear in several peer-reviewed international journals over the past few years, which relate to *the impact* of haze episodes on the air quality in the northern region of Thailand (Kim et al., 2011; Wiwatanadate and Liwsrisakun, 2011; Li et al., 2012). For instance, one study observed a 20% rise in the number of patients suffering from respiratory diseases in March 2007 when compared to one year earlier in 2006 (Pengchai et al., 2009). This can be correlated with the increase in PM_{10} levels caused by large-scale '*hot spots*,' locations and/or activities identified as high-density sources of pollutants, including the burning of agricultural waste, forest fires and trans-boundary haze pollution. The city of Chiang Mai is the largest in Northern Thailand, with a population of 160,000. It is a tourist attraction and noteworthy for the preservation of its distinctive local "Lanna" traditions. Chiang Mai's climate is categorized as a tropical wet and dry climate based on The Köppen climate classifications, with moderately warm to hot weather year-round, while night time temperatures are much lower than daytime

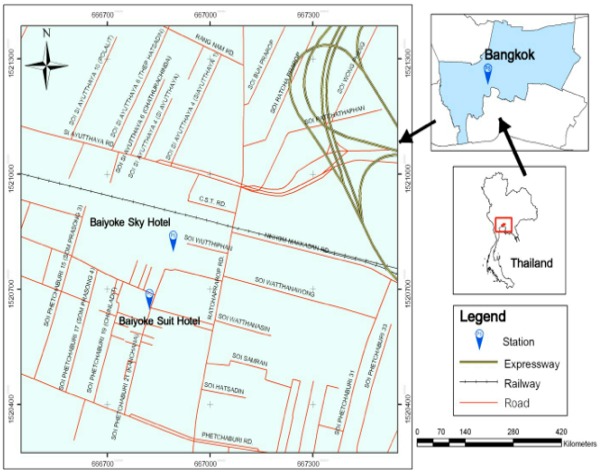

Figure 1. Description of Air Sample Observatory Sites at Bangkok

during the dry season.

Notably, two of the sampling sites — the Centara Duangtawan Hotel (18°47'03.46" N 98°59'56.72" E) and the Imperial Mae Ping Hotel (18°46'59.03" N 98°59'53.29" E) —were selected because they are located in the most densely populated area of the city, surrounded by bustling street markets, including the well-known "Night Bazaar." All three monitoring sites were also selected according to their altitude. Site-1 and Site-3 were situated at the Centara Duangtawan Hotel at 12 m and 152 m above ground level respectively, whilst Site-2 was established at the Imperial Mae Ping Hotel at 52 m above the building basement. Intensive monitoring campaigns were performed at all sites simultaneously, PM_{10} samples were collected every three hours consecutively from 21:00 Monday the 25th to 21:00 Thursday the 28th February 2008 using the Andersen High Volume Air Sampler PM_{10} TE6001 with a flow rate of 1.132 m^3 min^{-1}. 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".

Hat-Yai: It is well known that Hat-Yai is the largest metropolitan area in Southern part of Thailand with a population of 157,359 (2008) in the city itself, occupying an area of 7,393 square kilometers on the eastern side of the Malaysian Peninsula. Hat-Yai is a business city located in the southern part of Thailand adjacent to the Malaysian border, approximately 30 km away in the south of Gulf of Thailand. Hat-Yai is the largest city of Songkhla Province, the largest metropolitan area in Southern, and third largest metropolitan area of the country. Although the air quality levels in Hat-Yai are categorized as satisfactory in normal condition, the atmospheric concentrations of air pollutants exceeded the national air quality standard as a consequence of trans boundary pollution from uncontrollable forest fire in Indonesia (Quah, 2002). Hat-Yai city is categorized as tropical monsoon climate (Type Am) based on The Köppen climate classifications, with the 30 year average highest and lowest recorded temperature of 32.3°C (90.1°F) and 22.9°C (73.2°F) respectively. The 30-year average precipitation was recorded as 134.5 mm. Two observatory stations, namely Novotel Centara Hat-Yai Hotel (7°00'20.65" N 100°28'15.65" E) and Lee

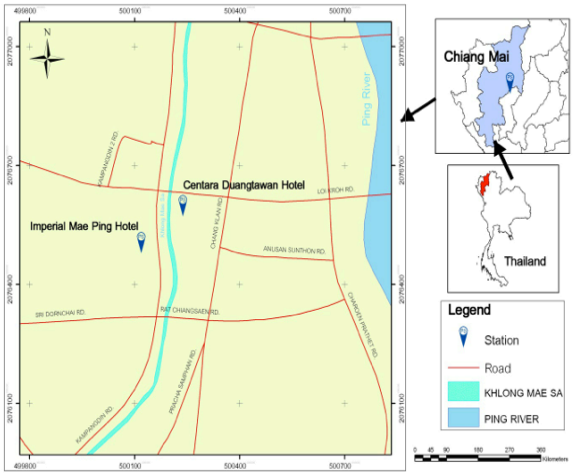

Figure 2. Description of Air Sample Observatory Sites at Chiang-Mai City

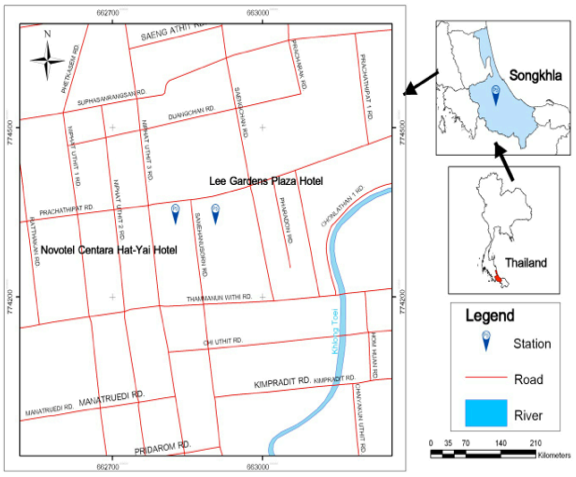

Figure 3. Description of Air Sample Observatory Sites at Hat-Yai City

Gardens Plaza Hotel (7°00'21.39" N 100°28'15.94" E), were located in the center of Hat-Yai city encompassed by schools, business buildings including cloth and snack shops in Kim Yong market. Three different measurement sites were prudently selected according to its elevation. Site-1 was placed at Novotel Centara Hat-Yai Hotel at 30 m above building basement, whilst Site-2 and Site-3 were positioned at Lee Gardens Grand Plaza Hotel at 60 m and 125 m above the ground level respectively. Intensive monitoring campaigns were performed at all measurement sites concurrently from December 17th to 20th 2007 in winter. PM_{10} samples were collected every three hours consecutively from 21:00 h December 17th to 21:00 h December 20th by using Graseby-Andersen High Volume Air Sampler. More detailed description of the air sampling method was provided in "Compendium Method IO-2.2. Sampling of Ambient Air for PM_{10} Using an Andersen Dichotomous Sampler".

Filter sample collection and meteorological data

PM_{10} High Volume Air Sampler: In 1971, The Environmental Protection Agency (EPA) promulgated primary and secondary national ambient air quality standards for particulate matter measured as "total suspended particulate" or "TSP". In 1987, the EPA announced changes in the standards to replace TSP as the indicator for particulate matter for the ambient standards by a new indicator that includes only those particles with an aerodynamic diameter less than or equal to nominally ten 10 μm (PM_{10}). In accordance with the USA Clean Air Act, high volume air samplers are the most widely utilized active air samplers for the collection of both particle-bound and vapour phase constituents of ambient air like carbonaceous aerosols. The sampler can draw large volumes of air (usually in the range of 800-1000 m^3) by suction through a 20cm x25cm filters (usually glass fibre, quartz tissue, or Teflon coated glass fibre filters) and adsorbent behind the filter to retain the vapour phase component. The selection of PM_{10} is performed by internal separation, followed by filtration and gravimetric determination of PM_{10} mass on the filter substrate.

In this research, a PM_{10} high volume air sampler TE-6001 (Tisch Environmental, Inc) and Variable Resistance

Calibration Kit TE-5028 were employed to collect air samples and measure the flow rate. Flow control is accomplished by restricting and thus accelerating the air-flow through the venturi tubes. At some point in the flow stream, the air velocity will equal the acoustic velocity and critical flow will be achieved. As long as downstream changes are small, all conditions at the venturi tubes (including the flow rate) are determined by upstream conditions. Since critical flow through the tubes are not greatly affected by changes in the filter loading, ambient temperature or barometric pressure, a stable volumetric flow rate is maintained as long as power is provided to the sampler blower motor. In contrast to passive air samplers, high volume air samples are relatively obtrusive, expensive and maintenance-intensive. However, the main advantages of high volume air samplers are their abilities to distinguish between particulate and vapour phase of carbonaceous aerosols coupled with affording opportunities to study relatively short-term temporal variation in concentrations.

Collecting Particulate OC/EC: Several factors govern the sampling artifact in both particulate and gaseous OC/EC in ambient air. The *ad*sorption of gaseous OC/EC onto a filter can cause positive biases in the measured particle-phase concentrations, and negative biases in the measured gas-phase concentrations. In order to investigate the sampling artifact effect, gas/particle partitioning can be parameterised using the coefficient K_p (m^3 μg^{-1}):

$$K_p=c_p/c_g \quad \text{Equation 8}$$

Where c_p is the measured particle-phase concentration (μg μg^{-1}) and c_g is the measured gas-phase concentrations (μg m^{-3}) respectively. The length of time over which sampling take place, the filter type of interest, the ambient temperature are the main factors which control the K_p values and therefore the sampling artifact (Mader and Pankow, 2000a; 2000b). To minimize the sampling artifact, the sampling time should be kept to a minimum to avoid fluctuations in temperature and concentration during sampling. The selection of filter type is also one of the most significant factors in controlling the measured gas/particle partitioning in ambient air. Mader and Pankow (2000a; 2000b) investigated the sampling artifact in Teflon membrane filters (TMFs) and quartz fiber filters (QFFs). It is well known that the TMFs have less surface area than the QFFs, thus the QFFs can be expected to be more absorptive than the TMFs. It was therefore assumed that the proportion of OC/EC adsorbed on QFFs are large compared to that adsorbed to TMFs (Mander and Pankow, 2000a; 2000b). By normalizing a gas/filter K_p value by the surface area af (m^2 g^{-1}) of the filter yields

$$K_{p,s}[m]=[Kp(m^3\mu g^{-1})]/[10^{-6}(g\mu g^{-1})\times a_f(m^2 g^{-1})] \quad \text{Equation 9}$$

Log $K_{p,s}$ values for OC/EC on QFFs were on average a factor of two higher than those for OC/EC found on TMFs. This result can also be explained by the larger surface area and higher energy surface of the QFFs resulting in a greater adsorptive affinity. The effects of temperature T (K) on the measured $K_{p,s}$ values were studied over the range 285-299 K. A 10º increase in T resulted in decreases in the $K_{p,s}$ values of PAHs, PCDFs and PCDDs by factors of 2.4, 3.1 and 3.4 respectively (Mader and Pankow, 2000). The relationship between $K_{p,s}$ and T displays by typical Van't Hoff behavior, and are correlated according to

$$logK_{p,s}=m_{p,s}/T+b_{p,s} \quad \text{Equation 10}$$

Values of $m_{p,s}$ and $b_{p,s}$ were unitless and evaluated using the common *y*-intercept regression (CYIR) method of Pankow (1991). The plots of log $K_{p,s}$ against log pLº (sub-cooled liquid vapour pressure) were found to be linear with slopes of approximately -1. At relative humidity (RH)=25% the pooled log $K_{p,s}$ data at 25ºC for the three compound classes were correlated with log pLº nearly as well (r^2=0.95) as were the data for the individual compound classes (r^2=0.97).

Organic carbon (OC) and elemental carbon (EC)

PM_{10} mass loadings were measured gravimetrically with a Mettler Toledo AB204-S electronic microbalance (Columbus, Ohio, USA). Prior to aerosol mass measurement, the QM/A were equilibrated for 24 h at a constant temperature between 20°C and 23°C and relative humidity between 35% and 45% in the laboratory of the Department of Civil and Structural Engineering, Research Centre for Urban Environmental Technology and Management, The Hong Kong Polytechnic University, China. Each filter was weighed at least three times before and after sampling following the 24-hour 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 attains its 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 IMPROVE TOC (Interagency Monitoring to Protect Visual Environments Total Organic Carbon) protocol (Chow et al., 1993; 2001; Fung et al., 2002). In addition, the quality control and quality assurance (QA/QC) protocols have been mentioned in detail in Cao et al. (2003).

Results

Chiang-Mai

OC/EC Concentrations in Chiang-Mai: Average and standard deviation values of PM_{10}, TC, OC and EC collected during the monitoring period in Chiang Mai are

illustrated in Table 1. At all sampling heights, the average concentrations for PM_{10} ranged from 49.4-203.1 μg m^{-3} and for TC, OC and EC from 2.6-70.0 μg m^{-3}, 2.6-60.0 μg m^{-3} and N.D. to 12.9 μg m^{-3}, correspondingly. At Site-1 (12 m above ground level), OC concentrations ranged from 2.6-51.0 μg m^{-3} with an average of 30.8±13.6 μg m^{-3}, contributing on average, 32.8±9.6% to the total PM_{10}. EC concentrations varied from N.D. to 10.9 μg m^{-3} with an average of 5.3±2.6 μg m^{-3}, contributing on average, 5.6±2.0% to the total PM_{10}. These results indicate that organic carbon is the most significant contributor to the total PM_{10} mass concentration. As illustrated in Table 2, the average OC concentration present in the samples taken in Chiang Mai (30.8 μg m^{-3}) is not only the highest on record, but more than three times greater than the highest followed by Zurich-Wiedikon, Switzerland (9.0 μg m^{-3}) and Bern, Switzerland (8.9 μg m^{-3}), while average EC concentrations were the highest in Hong Kong Polytechnic University, China (13.5 μg m^{-3}), followed by Zurich-Wiedikon, Switzerland (7.7 μg m^{-3}) and Bern, Switzerland (5.6 μg m^{-3}), respectively. The EC concentrations for Chiang Mai (5.3 μg m-3) were within the same range as those of Bern, Switzerland (5.6 μg m-3) and Guangzhou, China (4.0 μg m^{-3}).

Although the vertical distribution of carbonaceous compositions collected in different levels were similar to each other, some significant differences in atmospheric concentration were observed in EC compositions. As illustrated in Table 1, the average concentrations of EC were 5.3±2.6, 6.8±2.2 and 5.4±2.3 for Site-1, Site-2 and Site-3 respectively. In spite of the fact that all monitoring site positions were adjacent to each other, the mean concentration of EC observed at Site-2 was significantly ($p<0.05$) 1.3 times higher than those detected at Site-1 and Site-3. These differences seem to correspond with a difference in the pattern of wind direction distribution. As illustrated in Figure 4, the wind direction at Site-1 was mainly a combination of East and East-Northeast (40%). Because Site-1 was located west of the Night Bazaar's main street, the consequences here reflect the major contribution made by traffic emissions. Conversely, the wind directions at Site-2 were predominantly a mixture of South-Southeast and South (28%) coupled with North-Northwest (16%), thus altering both the air mass origin and carbonaceous composition to a certain extent. Interestingly, the wind direction distribution pattern at Site-2 was found to have considerable similarities with that at Site-3 suggesting that Southerly and Northwesterly wind directions correspond to an air parcel which has travelled at least for some time over agricultural waste burning areas. For OC, the differences in PM_{10} between Site-1 vs. Site-2 and Site-1 vs. Site-3 were not obvious in most samples.

Diurnal Variation of Carbonaceous Aerosols in Chiang-Mai: Three-hour-block averages of OC and EC concentrations were coupled with meteorological records to investigate potential trends in the diurnal and vertical variability of carbonaceous aerosols. Figure 5 shows diurnal variation of OC and EC concentrations in PM_{10} at different heights from February 25th to 28th 2008. Both OC and EC exhibit marked peaks during the heavy morning traffic (0900-1200 h) and the night rush hours (2100-2400 h) at all sampling heights. On the first day, the OC and EC curves appear to increase in concentration levels over the course of the morning, i.e. from 03:00-06:00 to 09:00-12:00, and then drop to lower concentrations later during the period of 12:00-15:00. The morning OC and EC peaks are highly pronounced as a consequence of the high volume of traffic and human activity, whilst the afternoon dip in mass concentrations of these carbonaceous aerosols can be attributed to the dilution effect caused by a thorough mixing with the daytime layer. In addition, the elevated concentrations of carbonaceous aerosols are not caused by only vehicular emissions but also agricultural waste burning in suburban regions.

As previously mentioned, uncontrolled biomass burning has been found to be a significant source of ambient PM_{10} in Chiang Mai and other northern cities over the past decades, particularly in the cold season. Since continuous biomass burning has no specific diurnal features, it seems reasonable to attribute both the morning and night peaks to the urban traffic emissions. This idea is strongly supported by the relatively high correlation coefficients (R) between OC and EC observed at Site-1 (R=0.94), Site-2 (R=0.74) and Site-3 (R=0.85). Another feature that may be deduced from Figure 5 is that the last day peaks of OC and EC were observed at Site-3 between 18:00 and 21:00 while the secondary peak appears between 12:00 and 15:00 at the same level. This trend is the reverse of carbonaceous diurnal variation observed at Site-1 and Site-2 during the same period. Since South-Southwesterly wind directions play a major role at Site-3 whilst East-Northeasterly and South-Southeasterly dominate the air parcel at Site-1 and Site-2 respectively, it is safe to assume that the long range transport of biomass burning particles has an tangible effect the air quality of

Table 1. Statistic Description of Concentrations of Carbonaceous Species and Its Percentage Contribution in PM_{10} Collected at Chiang-Mai, Bangkok and Hat-Yai

Sampling Height (m)	PM_{10} (μg m^{-3})	TC (μg m^{-3})	OC (μg m^{-3})	EC (μg m^{-3})	TC (%)	OC (%)	EC (%)
Chiang-Mai-Site-3	91.9±30.1	38.2±13.2	32.7±11.0	5.4±2.3	43.1±15.6	37.0±13.1	6.1±2.7
Chiang-Mai-Site-2	110.3±33.1	40.4±12.5	33.7±10.5	6.8±2.2	37.4±8.0	31.1±6.9	6.3±1.3
Chiang-Mai-Site-1	92.3±28.10	36.1±16.1	30.8±13.6	5.3±2.6	38.4±11.5	32.8±9.6	5.6±2.0
Bangkok-Site-3	112.3±18.0	84.0±15.8	68.4±13.2	15.6±3.5	75.3±10.8	61.3±9.8	14.0±2.0
Bangkok-Site-2	113.0±18.6	87.3±21.6	68.8±16.9	18.5±5.5	77.5±14.1	61.0±10.9	16.4 ±4.1
Bangkok-Site-1	118.7±21.1	81.2±19.6	62.9±15.0	18.4±5.3	68.4±10.8	53.0±9.2	15.3±2.7
Hat-Yai-Site-3	46.6±14.1	4.9±1.3	4.8±1.3	0.1±0.1	12.5±8.6	12.3±8.6	0.2±0.3
Hat-Yai-Site-2	41.2±5.9	5.7±1.9	5.4±1.6	0.4±0.3	14.2±4.5	13.3±3.9	0.9 ±0.8
Hat-Yai-Site-1	42.5±11.0	8.8±3.1	8.3±2.5	0.5±0.8	23.4±14.7	21.8±12.0	1.6±3.3

Table 2. Average OC and EC Concentrations and the OC/EC Ratios in PM_{10} Collected at Various Urban Atmospheric Environments

Sampling site	Area type	Above ground	Period	OC ($\mu g\ m^{-3}$)	EC ($\mu g\ m^{-3}$)	OC/EC
Chiang-Mai, Thailand[a]	Urban	12 m	February, 2008	30.8±13.6	5.3±2.6	5.8±3.9
Bangkok, Thailand[a]	Urban -background	38 m	February, 2008	62.9±15.0	18.4±5.3	3.4±1.3
Hat-Yai, Thailand[a]	Urban	30 m	December, 2007	8.3±2.5	0.5±0.8	16.2±26.2
Guangzhou, China[b]	Urban	12 m	August, 2006 – August, 2007	7.1±3.3	4.0±2.5	1.8±1.4
Zhaoqing, China[b]	Semi-rural	12 m	August, 2006 – August, 2007	8.2±5.0	3.9±2.0	2.1±1.7
Hong Kong, Polytechnic University, China[b]	Roadside	1.5 m	August, 2006 – August, 2007	7.9±3.8	13.5±3.5	0.6±0.3
Hong Kong, Hok Tsui, China[b]	Rural	1.5 m	August, 2006 – August, 2007	4.1±3.0	1.8±1.2	2.3±2.3
Whitbourne, UK[c]	Rural	1.5 m	November – December, 2003	3.2±1.5	1.1±0.5	2.9±1.9
Morogoro, Tanzania[d]	Rural	6 m	March-April, 2006	4.5±0.9	0.52±0.16	8.7±3.2
Zurich-Wiedikon, Switzerland[e]	City Center	1.5 m	July/August, 1998 and January/ February, 1999			
Zurich-Kaserne, Switzerland[e]	City Center	1.5 m	April, 1998-March, 1999	4.9	2.0	2.5
Basel, Switzerland[e]	Suburban	1.5 m	April, 1998-March, 1999	4.7	1.8	2.6
Payerne, Switzerland[e]	Rural	1.5 m	May/June and September/ October, 1999	3.4	1.3	2.6
Bern, Switzerland[e]	City Center	1.5 m	April, 1998-March, 1999	8.9	5.6	1.6
Chaumont, Switzerland[e]	Rural	1.5 m	April, 1998-March, 1999	1.7	0.6	2.8
Sihwa area, South Korea[f]	Urban-industrial	20 m	February, 1998-February, 1999	9.8±6.3	1.8±1.6	5.4±6.0
Houtem, Belgium[g]	Rural-background	1.5 m	September, 2006-September, 2007	3.2	0.5	6.8
Zelzate, Belgium[g]	Industrial site	1.5 m	September, 2006-September, 2007	4.4	1.3	3.4
Mechelen, Belgium[g]	Suburban-background	1.5 m	September, 2006-September, 2007	5.1	1.3	3.9
Thessaloniki, Greece[h]	Urban-industrial	1.5 m	December, 2006-March, 2007	8.7	2.9	3.0
Thessaloniki, Greece[h]	Urban-traffic	3.0 m	December, 2006-March, 2007	8.1	1.8	4.4
Cheju, Korea[i]	Urban-traffic	1.5 m	June-August, 1994	3.7	0.3	12.3
Kosan, Korea[i]	Rural-background	1.5 m	July-August, 1994	4.6±0.5	0.4±0.06	12.1±2.3
Seoul, South Korea[j]	Urban-background	20 m	June, 1999	10.3±3.2	8.4±3.0	1.2±0.6
Sihwa area, South Korea[k]	Urban-industrial	20 m	February, 1998-February, 1999	9.8±6.3	1.8±1.6	5.4±6.0
Hong Kong, China[l]	Urban-traffic	1.5 m	January-February, 2002	10.5±4.0	5.1±2.7	2.1±1.3
Guangzhou, China[l]	Urban-background	15 m	January-February, 2002	29.4±22.2	10.4±6.8	2.8±2.8
Shenzhen, China[l]	Urban-background	8 m	January-February, 2002	16.4±5.3	7.3±2.0	2.2±1.0
Borgerhout, Belgium[m]	Urban	1.5 m	September, 2006-September, 2007	5.3	2.0	2.6
Aarschot, Belgium[m]	Rural	1.5 m	September, 2006-September, 2007	4.1	1.0	4.2
Hasselt, Belgium[m]	Suburban	1.5 m	September, 2006-September, 2007	4.3	1.2	3.6
Thessaloniki, Greece[n]	Urban-industrial	1.5 m	June-September, 2007	6.4	2.9	2.2
Thessaloniki, Greece[n]	Urban-traffic	3.0 m	June-September, 2007	7.7	2.6	2.9
Kosan, Korea[o]	Rural	1.5 m	January, 1997	3.3±0.6	0.2±0.04	14.4±3.5
Chongju, South Korea[p]	Urban-Industrial	1.5 m	October, 1995-August, 1996	5.0±2.4	4.4±2.7	1.1±0.9
Beijing, China[q]	Downtown	1.5 m	July-September, 1999	21.5	8.7	2.5
Zhuhai[r]	Urban-background	20 m	January-February, 2002	14.5±4.6	6.0±1.8	2.4±1.1
Xi'an, China[s]	Urban-background	10 m	September-October, 2003	43.2±27.1	15.0±10.7	2.9±2.7
Xi'an, China[s]	Urban-background	10 m	November, 2003-February, 2004	93.0±58.4	22.7±12.3	4.1±3.4
Tongliao, China[t]	Rural-background	12 m	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. (2001), [l]Cao et al. (2003), [m]Vercauteren et al. (2011), [n]Terzi et al. (2010), [o]Kim et al. (2000), [p]Lee and Kang (2001), [q]He et al. (2001), [r]Cao et al. (2003), [s]Cao et al. (2005), [t]Li et al. (2006)

sampling site at Site-3.

Bangkok

<u>*OC/EC Concentrations in Bangkok*</u>: Statistical description of PM_{10}, carbonaceous compositions along with its percentage contributions in each sampling height is displayed in Table 1. PM_{10} mass concentrations at level-1 ranged from 88.4-164.6 $\mu g\ m^{-3}$, and at level-2 they ranged from 78.1-156.6 $\mu g\ m^{-3}$ whilst at level-3 they varied from 81-175.7 $\mu g\ m^{-3}$. The average concentrations of TC at level-1, level-2 and level-3 were 81.2±19.6 $\mu g\ m^{-3}$, 87.3±21.6 $\mu g\ m^{-3}$ and 84.0±15.8 $\mu g\ m^{-3}$ individually (Table 1). In the similar pattern, OC concentrations at level-1, level-2 and level-3 ranged from 38.5-88.6 $\mu g\ m^{-3}$, 48.8-108.1 $\mu g\ m^{-3}$ and 43.6-92.6 $\mu g\ m^{-3}$, respectively, with average mass percentage contribution of 53.0±9.2%, 61.0±10.9% and 61.3±9.8 in that order. The average EC concentration was significantly lower ($p<0.05$) in level-3 (15.6±3.5 $\mu g\ m^{-3}$) in comparison with those detected at level-1 (18.4±5.3 $\mu g\ m^{-3}$) and level-2 (18.5±5.5 $\mu g\ m^{-3}$). It is also interesting to note that the lowest average percentage contribution of EC in PM_{10} was also observed in level-3 (14.0±2.0%). On average, the atmospheric concentrations of OC explained 53.0± 9.2%, 61.0 ±10.9% and 61.3±9.8% of the total PM_{10} mass, determined gravimetrically at level-1, level-2 and level-3 in that order. As illustrated in Table 2, the highest OC concentrations were in Bangkok (62.9±15.0 $\mu g\ m^{-3}$), followed by Guangzhou, China (29.4±22.2 $\mu g\ m^{-3}$) and Shenzhen, China (16.4±5.3 $\mu g\ m^{-3}$), while average EC concentrations were also the highest in Bangkok (18.4±5.3 $\mu g\ m^{-3}$), followed by Guangzhou, China (10.4±6.8 $\mu g\ m^{-3}$) and Seoul, South Korea (8.4±3.0 $\mu g\ m^{-3}$), respectively.

<u>*Diurnal Variation of Carbonaceous Aerosols in Bangkok*</u>: Since Baiyoke Suit Hotel and Baiyoke Sky Hotel Baiyok Tower are located in "Pratunam (or Watergate in Thai)", one of the largest night market area in Thailand, it seems rational to consider the daily OC and EC mass concentration maximum at 2100-0000 as a consequence of nighttime traffic emission. A lower nocturnal mixing

Table 3. Statistical description of the incremental lifetime particulate matter exposure (*ILPE*) of PM_{10}, total carbon (TC), organic carbon (OC) and elemental carbon (EC) at Chiang-Mai, Bangkok and Hat-Yai, Thailand

	PM_{10} [g]		TC [g]		OC [g]		EC [g]	
	Male	Female	Male	Female	Male	Female	Male	Female
Chiang-Mai-Site-3	3.067±1.005	1.689±0.553	1.275±0.441	0.702±0.243	1.091±0.367	0.601±0.202	0.180±0.077	0.099±0.042
Chiang-Mai-Site-2	3.681±1.105	2.027±0.608	1.348±0.417	0.742±0.230	1.125±0.350	0.619±0.193	0.227±0.073	0.125±0.040
Chiang-Mai-Site-1	3.081±0.938	1.696±0.516	1.205±0.537	0.663±0.296	1.028±0.454	0.566±0.250	0.177±0.087	0.097±0.048
Bangkok-Site-3	3.748±0.601	2.064±0.331	2.804±0.527	1.544±0.290	2.099±0.441	1.156±0.243	0.614±0.117	0.338±0.064
Bangkok-Site-2	3.771±0.621	2.076±0.342	2.914±0.721	1.604±0.397	2.296±0.564	1.264±0.311	0.617±0.184	0.340±0.101
Bangkok-Site-1	3.962±0.704	2.181±0.388	2.710±0.654	1.492±0.360	2.099±0.501	1.156±0.276	0.614±0.177	0.338±0.097
Hat-Yai-Site-3	1.555±0.471	0.856±0.259	0.164±0.043	0.090±0.024	0.160±0.043	0.088±0.024	0.003±0.003	0.002±0.002
Hat-Yai-Site-2	1.375±0.197	0.757±0.108	0.190±0.063	0.105±0.035	0.180±0.053	0.099±0.029	0.013±0.010	0.007±0.006
Hat-Yai-Site-1	1.418±0.367	0.781±0.202	0.294±0.103	0.162±0.057	0.277±0.083	0.153±0.046	0.017±0.027	0.009±0.015
Average	2.851±1.095	1.570±0.603	1.434±1.131	0.789±0.623	1.151±0.853	0.634±0.469	0.274±0.269	0.151±0.148

*Department of health 2005 (Average Body Weight); Male: 58.25±9.76 kg; Female: 54.95±10.48 kg. Life Expectancy; Source: National Statistical Office Thailand, 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 $m^3 h^{-1}$

Table 4. Statistical Description of the Inhaled Particulate Mass of PM_{10}, Total Carbon (TC), Organic Carbon (OC) and Elemental Carbon (EC) at 15 Sites in Songkhla Province, Thailand[a]

Site	PM_{10} [g][b]		TC [g][b]		OC [g][b]		EC [g][b]	
	Male	Female	Male	Female	Male	Female	Male	Female
PSU1	1.191±0.344	0.656±0.189	0.223±0.280	0.123±0.154	0.161±0.189	0.089±0.104	0.061±0.029	0.034±0.016
PSU2	0.930±0.291	0.512±0.160	0.163±0.121	0.090±0.067	0.119±0.065	0.066±0.036	0.044±0.022	0.024±0.012
TI	1.567±1.021	0.863±0.562	0.495±1.250	0.273±0.688	0.286±0.353	0.158±0.194	0.209±0.460	0.115±0.253
CI	1.199±0.957	0.660±0.527	0.250±0.531	0.138±0.293	0.175±0.195	0.096±0.108	0.075±0.136	0.041±0.075
CPF	0.818±0.179	0.451±0.099	0.246±0.515	0.136±0.283	0.172±0.148	0.095±0.082	0.074±0.141	0.041±0.078
SL1	0.461±0.096	0.254±0.053	0.143±0.067	0.079±0.037	0.102±0.048	0.056±0.026	0.041±0.025	0.023±0.005
SL2	0.388±0.123	0.214±0.068	0.033±0.062	0.018±0.034	0.025±0.013	0.014±0.007	0.007±0.013	0.004±0.007
RMF1	1.147±0.287	0.632±0.158	0.528±1.057	0.291±0.582	0.362±0.573	0.199±0.315	0.166±0.203	0.091±0.112
RMF2	1.224±0.525	0.674±0.289	0.368±0.827	0.203±0.455	0.231±0.349	0.127±0.192	0.137±0.228	0.075±0.126
BT	1.430±0.830	0.787±0.457	0.471±0.981	0.259±0.540	0.269±0.272	0.148±0.150	0.202±0.368	0.111±0.202
WI	2.889±2.184	1.590±1.203	0.816±2.165	0.449±1.192	0.181±0.376	0.100±0.207	0.635±1.049	0.350±0.578
BF	1.009±0.470	0.556±0.259	0.210±0.294	0.115±0.162	0.162±0.087	0.089±0.048	0.048±0.062	0.026±0.034
PR	0.837±0.307	0.461±0.169	0.322±0.764	0.177±0.421	0.182±0.290	0.100±0.160	0.141±0.246	0.077±0.136
KHH	0.319±0.132	0.176±0.073	0.062±0.041	0.034±0.022	0.045±0.030	0.025±0.016	0.017±0.002	0.009±0.001
RSB	7.267±3.207	4.001±1.766	2.682±3.068	1.477±1.689	2.169±1.735	1.194±0.955	0.513±0.420	0.282±0.231
BB	0.851±0.076	0.469±0.042	0.371±0.336	0.204±0.185	0.266±0.075	0.147±0.041	0.104±0.090	0.057±0.050
PTB	2.792±0.776	1.537±0.427	1.615±2.219	0.889±1.222	1.288±1.173	0.709±0.646	0.327±0.337	0.180±0.185

*_Prince of Songkla University_ (**PSU**): The site was situated at about 3 m above ground level in the Faculty of Environmental Management of Prince of Songkla University, and about 550 m away from the main traffic road that leads to the city center of Hat-Yai. It is important to note that PSU1 and PSU2 represent the sampling period of June (28th-30th June, 2007) and October (24th-26th October, 2007) respectively. This site is considered as an urban residential zone. _Traffic Intersection_ (**TI**): The station was located at the traffic intersection in front of the main gate of PSU. It is situated on the eastern side and approximately 2.5 km far away from the Hat-Yai city center. This station is regarded as a traffic area closed to urban residential zone. The air samples were collected on 5th-7th July, 2007. _Corpse Incinerator_ (**CI**): This station is a part of Kor-Hong temple, located at the northern side and about 1.5 km far way from TI. Since timbers and tires were generally used as fuel for corpse incineration, this site is considered as an emission source of both timbers and tires-burning. This site represents the sampling period of 19th-21st July 2007. _Charoen Phokphand Factory_ (**CPF**): This site was situated inside the fish can manufacturing factory of Charoen Phokphand group, which is the largest business conglomerate in Thailand. As crude oil was used for the fish can production, this station can be regarded as an emission source of crude oil-burning. The monitoring was conducted on 24th-26th July 2007. _Songkhla Lake_ (**SL**): This station was located at the south of Songkhla Lake and approximately 13 km far away from the northern side of PSU. This site is also situated about 14 km away from the western side of the Gulf of Thailand. Since there are not many industrial and/or traffic emission sources including chemical and metallurgy factories, power plants, and etc in this district, we consider this site as a rural background sampling station. SL1 and SL2 represent the monitoring period of July (27th-29th July, 2007) and October (20th-22nd October, 2007) respectively. _Rubber Sheet Manufacturing Factory 1_ (**RMF1**): This monitoring site was located at Tumbol Tungwan, Hat-Yai district. As a part of manufacturing process, the rubber sheet was treated with steam of high temperature and high pressure coupling with the purification by using sulfuric acid solution. Since Para rubber trees were used as fuel for this process, this site represents an emission of mixed Para rubber trees burning, latex fragments and sulfuric acid aerosols. The air samples were collected from 30th July to 1st August 2007. _Rubber Sheet Manufacturing Factory 2_ (**RMF2**): This station was situated at Tumbol Tachang, Banglum district. Both RMF1 and RMF2 are regarded as an emission of mixed Para rubber trees burning, latex fragments and sulfuric acid aerosols. The air samples were collected from 2nd-4th August 207. _Bus Terminal_ (**BT**): This site was located at the south-western side of PSU and approximately 1.4 km far away from the campus. This station was selected as a source of diesel emission because the majority of these buses are diesel-fueled. The air sample collection was started from 5th-7th August 2007. _Waste Incinerator_ (**WI**): This site was situated at the city center and belongs to the municipality of Hat-Yai city. Since the municipal waste incinerated is a heterogeneous mixture of solid wastes and burning fuels, this site can be recognized as a combination of solid waste burning and diesel exhaust emission. The air samples were collected from 28th-30th August, 2007. _Barbeque Festival_ (**BF**): This site was located inside the PSU campus on the top roof of Faculty of Natural Resources. The barbeque festival has become an annual tradition that is held on the second week in August. The 40th Annual Barbecue Festival is set for Wednesday, August 15th, 2007. This site can be considered as an emission of charcoal burning. The air samples were collected from 15th-18th August, 2007. _Petkrasem Road_ (**PR**): This station was located at the heart of Hat-Yai city. The air mass collected at this area reflected the heaviest burden from traffic congestions with the mixture of diesel and benzene exhaust emissions. The monitoring was conducted on 27th-29th August, 2007. _Kor-Hong Hill_ (**KHH**): This site was situated at the radio station on the top of Kor-Hong hill with the elevation of 356 meters. The air mass passed over the station was considered as a mixture of all emission sources in urban area, and thus can be regarded as an urban residential zone monitoring site. The sampling was conducted on 3rd-5th November, 2007. _Rice Straw Burning_ (**RSB**): The rice straw burning has been the major practice for removing rice straw because it is fast, economical and practical in removing disease organisms. Although the options for the disposition of rice straw are limited, this practice leads to unacceptable air pollution. The station was situated at rice field in Satingpra district, Songkhla Province and considered as a represent of biomass burning. The sampling was conducted on 16th November, 2007. _Biomass Burning_ (**BB**): As a part of soil preparation process, the biomass must be disposed of in order to make way for the plantation. The sampling site was adjacent to the unused land, and located at Namom district, Songkhla Province. This site can be regarded as an emission source of biomass burning. The sampling was conducted on 17th November, 2007. _Para Rubber Tree Burning_ (**PTB**): This station is located Namom district, Songkhla Province and can be recognized as an emission source of Para rubber tree burning. The air samples were collected on 18th November, 2007.

[a]Inhaled particulate mass over exposure duration of 25 years. [b]Values represent average±standard deviation

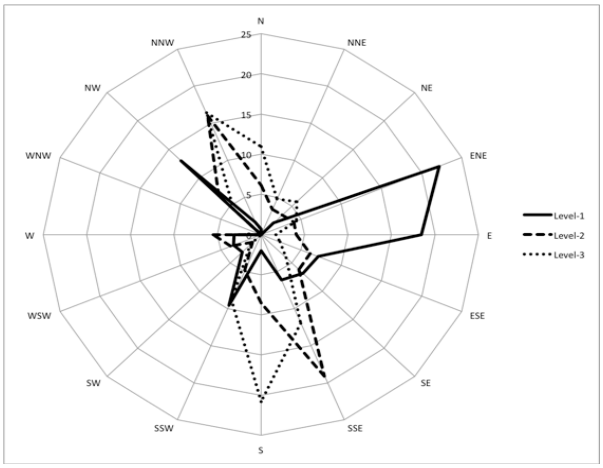

Figure 4. Description of Wind Rose in Chiang-Mai from 25/02/08-28/02/08

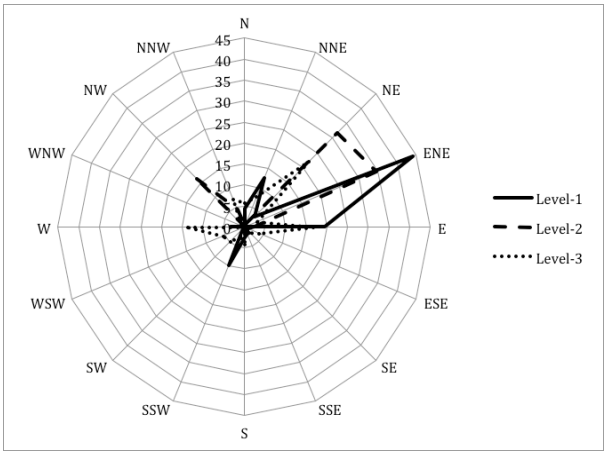

Figure 6. Percentage Contributions of Wind Directions at Air Sample Observatory Sites in Bangkok from 18/02/2008-21/02/2008

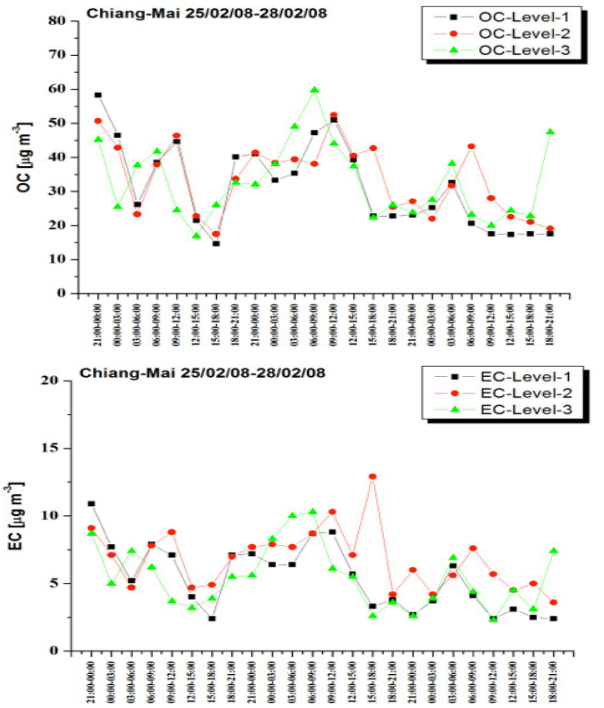

Figure 5. Diurnal Variation of OC and EC in PM_{10} Collected at Chiang-Mai from 25/02/08-28/02/08

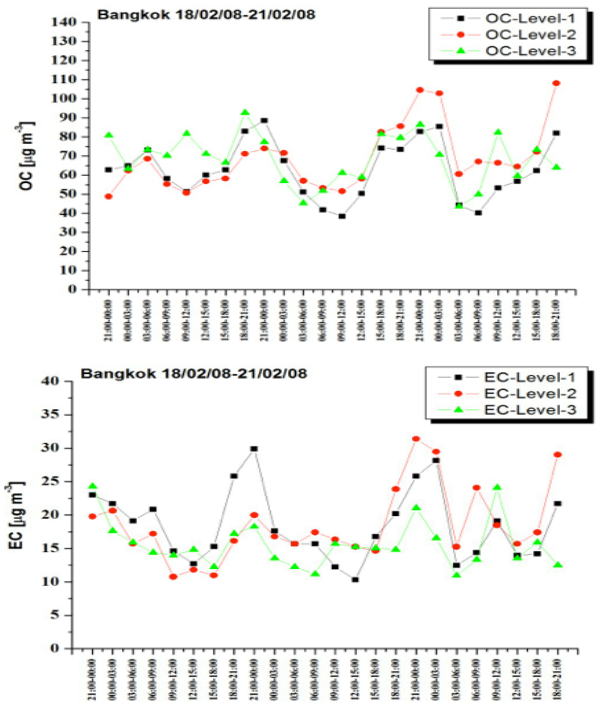

Figure 7. Diurnal Variation of OC and EC in PM_{10} Collected at Bangkok from 18/02/2008-21/02/2008

layer and a stable upper layer between 2100 and midnight can significantly enhance the atmospheric chemical compositions as well explained by Wilson and Stockburger (1990) and Beyrich (1997). The decreased wind speed around midnight can decelerate atmospheric dispersion and thus increase the carbonaceous aerosol concentrations during the monitoring period of 2100-0000. On the contrary, the midday minimum OC and EC concentrations can be attributed to several reasons. Firstly, the emission source strengths are lower during daytime in comparison with those of high traffic density at nighttime. Secondly, the height of developing daytime mixing layer enhances and mixed with the relatively clean air above, which triggers the dilution effect. Thirdly, the high daytime wind speed caused greater dispersion and thus decreasing the atmospheric concentration of carbonaceous aerosols.

As formerly mentioned, continuous vehicular exhaust has been assumed to be a major source of carbonaceous aerosols in Bangkok over the past decades, particularly at the heart of the metropolis. This assumption is strongly supported by the relatively high correlation coefficients (R) between OC and EC observed at Site-1 (R=0.81), Site-2 (R=0.79) and Site-3 (R=0.66). The decrease of R observed at Site-2 and Site-3 reflects the stronger influence of dilution effect at higher altitude. It is also worth to mention that wind direction can play an important role in governing the variation of carbonaceous particles particularly at higher altitude. For instance, the morning peaks of OC were detected at Site-3 at 0900-1200 during the three days of monitoring period. This opposed to those variation pattern of Site-1 and Site-2, which seems to decrease at the same observation phase. This phenomenon can be explained by numerous factors. Since East-Northeasterly and Northeasterly wind directions play a crucial role at Site-1 and Site-2 whilst Easterly and Westerly wind directions govern the majority of air

Table 5. Statistical Description of Gas-particle Partitioning Coefficient (K_p) of PAHs in PM_{10} Estimated by Dachs-Eisenreich Model

	Ac	Ace	Fl	Ph	An	Pyr
Chiang-Mai-Site-3	$2.04\times10^{-5}\pm3.84\times10^{-5}$	$2.88\times10^{-5}\pm5.76\times10^{-5}$	$6.43\times10^{-5}\pm1.46\times10^{-4}$	$3.27\times10^{-4}\pm9.56\times10^{-4}$	$3.54\times10^{-4}\pm1.05\times10^{-3}$	$3.15\times10^{-3}\pm1.28\times10^{-2}$
Chiang-Mai-Site-2	$1.99\times10^{-5}\pm1.79\times10^{-5}$	$2.81\times10^{-5}\pm2.68\times10^{-5}$	$6.26\times10^{-5}\pm6.81\times10^{-5}$	$3.18\times10^{-4}\pm4.44\times10^{-4}$	$3.44\times10^{-4}\pm4.87\times10^{-4}$	$3.05\times10^{-3}\pm5.95\times10^{-3}$
Chiang-Mai-Site-1	$1.85\times10^{-5}\pm2.20\times10^{-5}$	$2.60\times10^{-5}\pm3.31\times10^{-5}$	$5.80\times10^{-5}\pm8.41\times10^{-5}$	$2.94\times10^{-4}\pm5.49\times10^{-4}$	$3.18\times10^{-4}\pm6.02\times10^{-4}$	$2.82\times10^{-3}\pm7.37\times10^{-3}$
Bangkok-Site-3	$4.70\times10^{-5}\pm3.06\times10^{-4}$	$6.64\times10^{-5}\pm4.62\times10^{-4}$	$1.49\times10^{-4}\pm1.19\times10^{-3}$	$7.63\times10^{-4}\pm7.95\times10^{-3}$	$8.26\times10^{-4}\pm8.72\times10^{-3}$	$7.43\times10^{-3}\pm1.10\times10^{-1}$
Bangkok-Site-2	$5.17\times10^{-5}\pm4.65\times10^{-5}$	$7.28\times10^{-5}\pm7.02\times10^{-4}$	$1.62\times10^{-4}\pm1.80\times10^{-3}$	$8.22\times10^{-4}\pm1.21\times10^{-2}$	$8.90\times10^{-4}\pm1.32\times10^{-2}$	$7.87\times10^{-3}\pm1.67\times10^{-1}$
Bangkok-Site-1	$4.52\times10^{-5}\pm3.90\times10^{-4}$	$6.34\times10^{-5}\pm5.89\times10^{-4}$	$1.40\times10^{-4}\pm1.51\times10^{-3}$	$7.02\times10^{-4}\pm1.01\times10^{-2}$	$7.59\times10^{-4}\pm1.11\times10^{-2}$	$6.60\times10^{-3}\pm1.39\times10^{-1}$
Hat-Yai-Site-3	$6.50\times10^{-7}\pm1.22\times10^{-5}$	$9.13\times10^{-7}\pm1.84\times10^{-5}$	$2.03\times10^{-6}\pm4.77\times10^{-5}$	$1.02\times10^{-5}\pm3.24\times10^{-4}$	$1.10\times10^{-5}\pm3.56\times10^{-4}$	$9.68\times10^{-5}\pm4.58\times10^{-3}$
Hat-Yai-Site-2	$2.86\times10^{-6}\pm8.79\times10^{-5}$	$4.01\times10^{-6}\pm1.33\times10^{-4}$	$8.89\times10^{-6}\pm3.46\times10^{-4}$	$4.45\times10^{-5}\pm2.35\times10^{-3}$	$4.81\times10^{-5}\pm2.58\times10^{-3}$	$4.19\times10^{-4}\pm3.33\times10^{-2}$
Hat-Yai-Site-1	$1.15\times10^{-5}\pm1.25\times10^{-4}$	$1.62\times10^{-5}\pm1.89\times10^{-4}$	$3.57\times10^{-5}\pm4.91\times10^{-4}$	$1.78\times10^{-4}\pm3.33\times10^{-3}$	$1.93\times10^{-4}\pm3.66\times10^{-3}$	$1.67\times10^{-3}\pm4.71\times10^{-2}$
Average	$2.42\times10^{-5}\pm1.63\times10^{-4}$	$3.41\times10^{-5}\pm2.46\times10^{-4}$	$7.59\times10^{-5}\pm6.32\times10^{-4}$	$3.84\times10^{-4}\pm4.23\times10^{-3}$	$4.16\times10^{-4}\pm4.64\times10^{-3}$	$3.68\times10^{-3}\pm5.86\times10^{-2}$

	Fluo	B[a]A	B[a]P	Ind	D[a,h]A	B[g,h,i]P
Chiang-Mai-Site-3	$3.41\times10^{-3}\pm1.24\times10^{-2}$	$8.03\times10^{-2}\pm5.18\times10^{-1}$	$7.47\times10^{-1}\pm4.72$	4.61±3.64	6.37±5.30×10	6.00±4.84×10
Chiang-Mai-Site-2	$3.30\times10^{-3}\pm5.77\times10^{-3}$	$7.74\times10^{-2}\pm2.40\times10^{-1}$	$7.20\times10^{-1}\pm2.19$	4.44±1.69	6.12±2.45×10	5.77±2.24×10
Chiang-Mai-Site-1	$3.05\times10^{-3}\pm7.14\times10^{-3}$	$7.12\times10^{-2}\pm2.98\times10^{-1}$	$6.63\times10^{-1}\pm2.72$	4.08±2.10	5.63±3.05×10	5.30±2.79×10
Bangkok-Site-3	$8.00\times10^{-3}\pm1.05\times10^{-1}$	$1.92\times10^{-1}\pm4.65$	1.79±4.23	$1.11\pm3.33\times10^{+2}$	$1.54\times10\pm4.87\times10^{+2}$	$1.44\times10\pm4.44\times10^{+2}$
Bangkok-Site-2	$8.53\times10^{-3}\pm1.60\times10^{-1}$	$1.99\times10^{-1}\pm7.05$	1.85±6.41	$1.14\pm5.05\times10^{+2}$	$1.57\times10\pm7.38\times10^{+2}$	$1.48\times10\pm6.74\times10^{+2}$
Bangkok-Site-1	$7.19\times10^{-3}\pm1.34\times10^{-1}$	$1.63\times10^{-1}\pm5.86$	1.52±5.33	$9.21\pm4.19\times10^{+2}$	$1.27\times10\pm6.12\times10^{+2}$	$1.20\times10\pm5.59\times10^{+2}$
Hat-Yai-Site-3	$1.05\times10^{-4}\pm4.36\times10^{-3}$	$2.42\times10^{-3}\pm1.99\times10^{-1}$	$2.25\times10^{-2}\pm1.81$	$1.38\times10^{-1}\pm1.44\times10$	$1.89\times10^{-1}\pm2.11\times10$	$1.79\times10^{-1}\pm1.93\times10$
Hat-Yai-Site-2	$4.56\times10^{-4}\pm3.16\times10^{-2}$	$1.04\times10^{-2}\pm1.45$	$9.66\times10^{-2}\pm1.31$	$5.88\times10^{-1}\pm1.05\times10^{+2}$	$8.09\times10^{-1}\pm1.54\times10^{+2}$	$7.63\times10^{-1}\pm1.40\times10^{+2}$
Hat-Yai-Site-1	$1.82\times10^{-3}\pm4.49\times10^{-2}$	$4.10\times10^{-2}\pm2.05$	$3.82\times10^{-1}\pm1.86$	$2.32\pm1.49\times10^{+2}$	$3.19\pm2.18\times10^{+2}$	$3.01\pm1.98\times10^{+2}$
Average	$3.98\times10^{-3}\pm5.61\times10^{-2}$	$9.30\times10^{-2}\pm2.48$	$8.65\times10^{-1}\pm2.25$	$5.32\pm1.78\times10^{+2}$	$7.34\pm2.60\times10^{+2}$	$6.92\pm2.37\times10^{+2}$

Table 6. Statistical Description of $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$ in PM_{10} Collected at Chiang-Mai, Bangkok and Hat-Yai, Thailand

	Ac	Ace	Fl	Ph	An	Pyr
Chiang-Mai-Site-3	13.09±15.22	12.32±14.20	11.46±13.05	9.67±10.74	9.61±10.65	7.50±6.89
Chiang-Mai-Site-2	15.91±14.59	14.98±13.61	13.94±12.51	11.75±10.30	11.68±10.22	9.14±6.61
Chiang-Mai-Site-1	13.52±16.79	12.73±15.66	11.84±14.40	9.99±11.85	9.93±11.75	7.77±7.59
Bangkok-Site-3	18.04±20.33	16.98±18.94	15.79±17.40	13.30±14.28	13.22±14.17	10.26±8.99
Bangkok-Site-2	21.30±24.95	20.05±23.25	18.66±21.35	15.74±17.53	15.64±17.39	12.25±11.04
Bangkok-Site-1	23.21±27.10	21.86±25.25	20.36±23.19	17.19±19.04	17.09±18.89	13.52±12.01
Hat-Yai-Site-3	1.65±5.89	1.56±5.48	1.45±5.03	1.22±4.12	1.21±4.09	0.96±2.57
Hat-Yai-Site-2	5.88±14.35	5.54±13.36	5.15±12.26	4.35±10.05	4.33±9.97	3.42±6.25
Hat-Yai-Site-1	4.78±24.49	4.50±22.81	4.19±20.93	3.54±17.15	3.52±17.02	2.79±10.68
Average	15.62±7.41	14.62±6.91	13.50±6.37	11.21±5.26	11.13±5.22	7.79±3.61

	Fluo	Chry	B[a]A	B[a]P	Ind	D[a,h]A	B[g,h,i]P
Chiang-Mai-Site-3	7.80±8.40	5.78±5.95	3.16±2.20	3.00±2.08	2.50±1.70	8.45±5.70	2.50±1.69
Chiang-Mai-Site-2	9.49±8.05	7.30±5.70	3.86±2.11	3.66±2.00	3.05±1.63	10.32±5.48	3.05±1.63
Chiang-Mai-Site-1	8.07±9.27	5.98±6.56	3.29±2.42	3.12±2.29	2.60±1.87	8.80±6.29	2.60±1.87
Bangkok-Site-3	10.73±11.14	7.93±7.85	4.28±2.80	4.07±2.64	3.39±2.15	11.44±7.23	3.39±2.15
Bangkok-Site-2	12.72±13.67	9.42±9.64	5.18±3.44	4.92±3.25	4.10±2.64	13.86±8.88	4.10±2.64
Bangkok-Site-1	13.92±14.85	10.34±10.48	5.81±3.75	5.52±3.54	4.61±2.88	15.57±9.69	4.61±2.88
Hat-Yai-Site-3	0.99±3.21	0.73±2.26	0.41±0.79	0.39±0.74	0.32±0.60	1.09±2.03	0.32±0.60
Hat-Yai-Site-2	3.52±7.82	2.61±5.50	1.47±1.91	1.39±1.80	1.16±1.47	3.93±4.93	1.16±1.46
Hat-Yai-Site-1	2.87±13.35	2.13±9.39	1.20±3.27	1.14±3.09	0.95±2.51	3.22±8.43	0.95±2.50
Average	8.88±4.14	6.41±2.97	2.85±1.45	2.70±1.38	2.23±1.15	7.52±3.89	2.23±1.15

mass at Site-3, it appears reasonable to assume that this discrepancy may be caused by the difference in wind distribution pattern. The drop of anthropogenic activities in midday may reduce the carbonaceous content at Site-1 and Site-2 because the shops in the street were for the most part closed during the period of 0900-1200. In addition, sea breeze may also somehow be responsible for the short-range transportation of carbonaceous aerosols from Map Ta Phut industrial estate, which is located 185 km at the Southeastern part of Bangkok adjacent to the Gulf of Thailand.

Hat-Yai

OC/EC Concentrations in Hat-Yai: Average and range atmospheric concentrations of PM_{10} and carbonaceous compositions are displayed in Table 5. At all sampling heights the major contributor to PM_{10} mass was OC followed by EC. It is also worth to mention that OC and EC exhibited significantly higher concentration at level-1 (30 m) in comparison to those observed at level-2 (60 m) and level-3 (125 m) ($p<0.05$). OC ranged from 4.6-13.9 $\mu g\ m^{-3}$, 3.2-9.3 $\mu g\ m^{-3}$ and 2.9-7.9 $\mu g\ m^{-3}$ for level-1, level-2 and level-3, respectively. OC components dominated the PM_{10} profiles at all levels accounting for 21.8±12.0%, 13.3±3.9% and 12.3±8.6% of total PM_{10} mass at level-1, level-2 and level-3, correspondingly. As illustrated in Table 6, the highest OC concentrations were in Xi'an, China (93.0±58.4 $\mu g\ m^{-3}$ and 43.2±27.1 $\mu g\ m^{-3}$), followed by Beijing, China (21.5 $\mu g\ m^{-3}$) and Tongliao, China (15.7 $\mu g\ m^{-3}$), while average EC concentrations were also the

highest in Xi'an, China (22.7±12.3 μg m^{-3} and 15.0±10.7 μg m^{-3}), followed by Beijing, China (8.7 μg m-3) and Zhuhai, China (6.0±1.8 μg m^{-3}), respectively. The data of EC Hat-Yai (0.5±0.8 μg m^{-3}) were in the range between those of Kosan, Korea (0.2±0.04 μg m^{-3}) and Aarschot, Belgium (1.0 μg m^{-3}).

Diurnal variation of OC and EC in Hat-Yai: Diurnal variation of three-hourly PM_{10}, TC, OC, and EC are displayed in Figure 9 and represent monitoring period of three days. During this observation period, the correlation coefficients (*R*) between OC and EC were highest at Site-1 (*R*=0.80) followed by Site-2 (*R*=0.60) and Site-3 (*R*=0.070) respectively. The altitude-decreasing pattern of *R* can be explained by numerous factors. Firstly, the photolysis and heterogeneous chemical reactions with trace gaseous species can be interpreted for this consequence. Since there were no significant differences (p<0.05) between solar radiation measured at Site-1 (109.3±149.9 W m^{-2}) vs Site-2 (98.7±115.9 W m^{-2}) and Site-2 vs Site-3 (145.6±183.9 W m^{-2}), it would be safe to assume that photolysis activities play a minor role in oxidizing carbonaceous aerosols and thus altering OC-EC correlations at higher altitude. Secondly, wind speed might influence the correlations between OC and EC due to the fluctuations in magnitude of dispersion occurred in different altitudes. Significant differences of wind speed (p<0.001) found in Site-1 (2.5±0.55 m s^{-1}) vs Site-2 (3.5±0.94 m s^{-1}) and Site-2 vs Site-3 (1.6±0.46 m s^{-1}) support this idea. Thirdly, the strength of individual emission sources varied with wind direction. Hence, significant differences of wind direction (p<0.001) detected at Site-1 (252±17°) vs Site-2 (100±11°) and Site-2 vs Site-3 (152±50°) emphasize the importance of emission sources on variation of R-values.

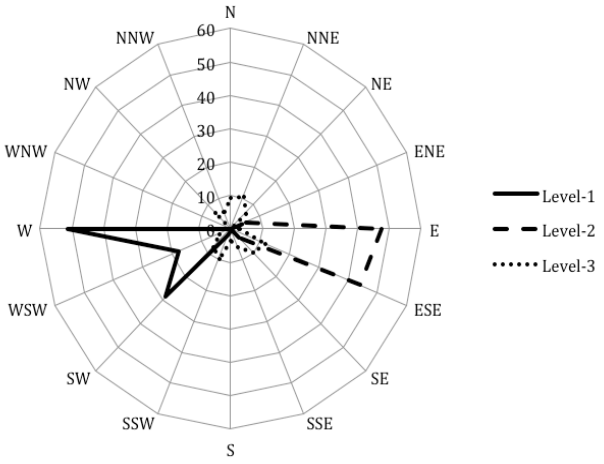

Figure 8. Percentage Contributions of Wind Directions in Hat-Yai from 17/12/07-20/12/07

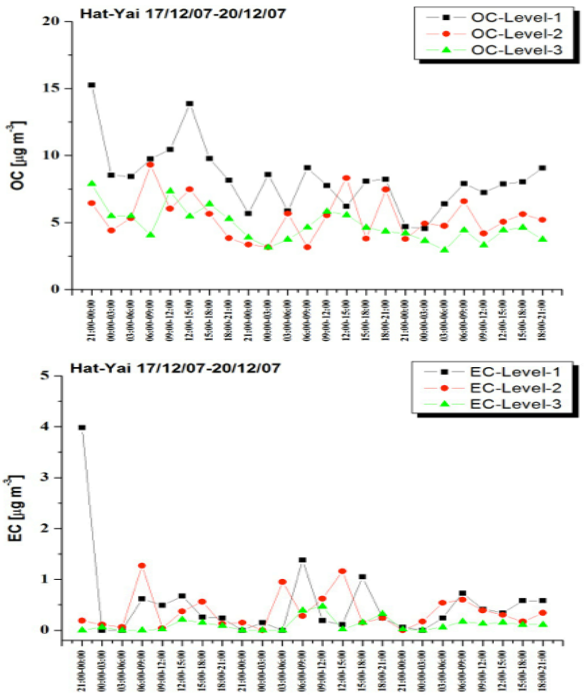

Figure 9. Diurnal Variation of OC and EC in PM_{10} Collected at Hat-Yai from 17/12/07-20/12/07

As illustrated in Figure 9, the maximum OC and EC concentrations were detected during the observation period of 21:00-00:00 on 17/12/2007 at Site-1 when Hat-Yai city showed the highest vehicular density at these peak hours. Since observatory sites are surrounded by restaurants and encompassed by night markets, it appears plausible to ascribe the highest content of carbonaceous aerosols from 21:00 to 00:00 as a mixture of traffic originated air pollutants. The overall declination of carbonaceous contents of all sampling sites was prominent after midnight of 17/12/2007. This can be explained by some small contributions of wet-depositions caused by irregular light shower rain occurred during the three days of monitoring campaign. The morning peak of OC and EC found during the sampling period of 06:00-09:00 reflected the crucial role of vehicular exhausts on carbonaceous contents in Hat-Yai city.

Discussion

Although several studies indicated that indoor PM_{10} concentrations, PAHs and VOCs were steadily lower than outdoor concentrations, the moderate positive correlations between indoor and outdoor concentrations were observed, suggesting that indoor concentrations were to some extent depending on ambient concentrations (Colome et al., 1992; Ohura et al., 2009; Masih et al., 2010). According to our best knowledge, there is no available study related to the assessment of health risk associated with the exposure to PM_{10}, OC and EC of workers and residents living in high buildings of Thailand. Hence, it is crucial to estimate the incremental lifetime particulate matter exposure (*ILPE*) by using the model, which can be defined as follows:

$$ILPE=C\times IR\times t\times EF\times ED \qquad \textbf{Equation 11}$$

Where: *ILPE*=incremental lifetime particulate matter exposure (g);

C=PM_{10}, OC and EC concentrations (μg m^{-3}); IR=Inhalation rate (m^3 h^{-1}); t=Daily exposure time span (6 hours d^{-1}, for two shifts); EF=Exposure frequency (250 d year^{-1} [a], upper-bound value); ED=Exposure duration (25 years[a], upper-bound value). Note: [a]Adapted from the Human Health Evaluation Manual (US EPA, 1991).

According to the methods for derivation of inhalation dosimetry (US EPA, 1994), the inhalation rates of male

and female outdoor 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's exposure to PAHs at work in China (Hu et al., 2007). The calculated *ILPE* levels in three different altitudes are summarized and compared with previous studies reported by Pongpiachan et al. (2009).

Interestingly, the estimated *ILPE* of the carbonaceous aerosols were constantly highest at Chiang-Mai-Site-2 and Bangkok-Site-2. For instance, the average values of TC accumulated in male and female workers in Bangkok and Chiang-Mai over a duration of 25 years were 2.914±0.721g, 1.604±0.397g and 1.348±0.417g, 0.742±0.230g respectively. This can be explained by several reasons. Firstly, the comparatively unique building configuration, orientation and surface area to volume (SA/V) ratio can dramatically govern the "heat transfer" and "airflow" of PM_{10} collected at Site-2. In other words, the building orientation determines the amount of radiation it receives. This will inevitably affect the expansion rate or air parcel at Site-2 and thus the air contents of carbonaceous aerosols. Secondly, wind speed and direction can be different at different altitudes. Without any doubts, these two meteorological parameters can noticeably change particulate OC/EC concentrations. Thirdly, the upwelling of air mass (i.e. a warmer, less-dense air mass moves over a cooler, denser air mass) from ground level brought more carbonaceous particles from traffic exhausts to Site-2. Fourthly, the dilution effect might have played a major role in reducing carbonaceous particles at Site-3.

In this study, the predicted *ILPE* of carbonaceous species were compared with other PM_{10} collected from various types of emission sources at Songkla province (Pongpiachan et al., 2009). Study area and description of sampling sites are clearly indicated in Table 4. The data set was categorized into seven groups according to its source characteristics (Group 1: PSU1, PSU2, urban residential zone; Group 2: SL1, SL2, KHH, rural background; Group 3: TI, BT, PR, traffic emissions; Group 4: CPF, RMF1, RMF2, industrial activities; Group 5: RSB, BB, PTB, biomass burnings; Group 6: CI, WI, incinerator emissions; and Group 7: BF, charcoal burnings). Interestingly, the average values of *ILPE* of TC at Baiyoke Suit Hotel and Baiyoke Sky Hotel are approximately ten times higher than those of PSU, CI, CPF and BF but slightly higher than those of RSB. This indicates a higher risk of developing lung cancer and other respiratory diseases across workers and residents living in high buildings located at Pratunam area than those who live adjacent to roads, factories and incinerators in Songkla province. In addition, the results highlight the risk for accumulation of carbonaceous particles at Baiyoke Suit Hotel and Baiyoke Sky Hotel, which is comparable to those of rice straw burning in Songkla province.

Estimation of gas-particle partitioning coefficients (K_p) and mechanisms of PAHs in various occupational environments: To obtain a quantitative understanding of the cancer risk, the gas-particle partitioning coefficients (K_p) of PAHs associated with different occupational environments were calculated according to Dachs-Eisenrich model (i.e. Equation 4). As illustrated in Table 5A and 5B, the highest K_p of all PAHs were in Chiang-Mai ($K_{P\text{-average}}$: 1.33; 1.85×10^{-5} ~ 6.73), followed by Bangkok ($K_{P\text{-average}}$: 3.22; 4.52×10^{-5} ~ 15.7) and Hat-Yai ($K_{P\text{-average}}$: 0.309; 6.50×10^{-7} ~ 3.19), While averaged $K_{P\text{-}D[a,h]A}$ was the highest ($7.34\pm2.60\times10^2$), followed by $K_{P\text{-}B[g,h,i]P}$ ($6.92\pm2.37\times10^2$) and $K_{P\text{-}Ind}$ ($5.32\pm1.78\times10^2$), respectively (Table 5A-5B). Because B[*a*]P is classified in Group II (probably carcinogenic to man), it is of great importance to conduct the sensitivity test relative to the highest $K_{P\text{-}B[a]P}$ of Bangkok. The concept of normalization is introduced based on consideration of $K_{P\text{-}B[a]P\text{-}Bangkok}/K_{P\text{-}B[a]P\text{-}other\ cities}$ ratio. The average ratios of $K_{P\text{-}B[a]P\text{-}Bangkok}/K_{P\text{-}B[a]P\text{-}other\ cities}$ of Chiang-Mai and Hat-Yai were 2.42 and 10.3 in that order. These results strongly suggest that regular exposure to harmful carcinogenic substances in Pratunam (Water Gate) area may be another leading cause of lung cancer raising concerns of long-term exposure of workers and residents to fine particulate PAHs in Bangkok in comparison with those of Chiang-Mai and Hat-Yai.

Recent work by Ribes et al. (2003) has demonstrated that the ratio of $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$ can be employed as an indicator to determine the dominant gas-particle partitioning. This ratio will allow clarification of the prevalent mechanisms for gas-particle partitioning of PAHs. Dachs and Eisenreich (2000) have suggested that the mechanisms governing gas-particle partitioning of PAHs can be considered into three scenarios or cases according to the ratio of $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$.

Case I: $(f_{EC}K_{SA}f_{OCT}/f_{OM}K_{OA})>5$

Case II: $5>(f_{EC}K_{SA}f_{OCT}/f_{OM}K_{OA})>0.2$

Case III: $(f_{EC}K_{SA}f_{OCT}/f_{OM}K_{OA})<0.2$

It has been assumed that Case I occurs when gas-particle partitioning dominated by *ad*sorption onto the soot phase, whereas Case III represents for the *ab*sorption into organic matter. On the other hand, Case II occurs when both *ab*sorption into organic layer and *ad*sorption onto the soot carbon is the major sorptive mechanism.

In this study, the ratios of $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$ ranging from 1.65-27.10 with the average of 15.62±7.41 for Ac, from 0.39-5.52 with the average of 2.70±1.38 for B[*a*]P, from 1.09-15.57 with the average of 7.52±1.02 for D[*a,h*]A and from 0.32-4.61 with the average of 2.23±1.15 for Ind respectively (Table 6A-6B). Based on the ratios of, it appears rationale to assume that the main gas-particle partitioning of B[b]F, B[a]P, Ind and B[g,h,i] P governing over the atmosphere of three cities is Case II (i.e. $0.2<f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}<5$), which indicates both *ab*sorption into organic layer and *ad*sorption onto the soot carbon (Table 6A-6B). This is in good agreement with earlier study highlighting that *ab*sorption into the organic matter is the dominant mechanism for gas-particle partitioning of PAHs on SOC (Liang et al., 1997). Previous studies indicated that *ad*sorption and *de*sorption of PAHs to combustion particles may take several hours to reach equilibrium (Strommen et al., 1999) and perhaps because of the existence of a liquid-like organic film coating the elemental carbon. Therefore the mass transfer rate

of PAHs is controlled by the diffusion from liquid-like organic phase into elemental carbon (Strommen et al., 1999). Nonetheless, the majority of PAHs namely Ac, Ace, Fl, Ph, An, Pyr, Fluo, Chry and D[*a,h*]A correspond to a $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$ ratio higher than 5 (Case I) and thus one can conclude that *ad*sorption completely governed gas-particle partitioning of low and medium molecular weight PAHs in the tree cities. Interestingly, it has been reported that *ab*sorption into organic matter may account for less than 10% of the total PAHs in Chesapeake Bay particulate phase (Dachs et al., 2000). In addition, it is worth mentioning that $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$ ratios of all PAHs are higher than 0.2 suggesting that *ab*sorption into organic layer alone might play a minor role in gas-particle partitioning of PAHs in Chiang-Mai, Bangkok and Hat-Yai. In spite of numerous efforts mentioned above to describe the prevailing gas-particle partitioning, it is still problematic to show an entirely clear picture of the relative importance of carbonaceous fractions in the gas-particle partitioning of the PAHs of interest here. For instance, the confounding effects of another sorptive compartment such as mineral surfaces may exist. The uncertainties in OC and EC determinations and measurement artifacts rule out some role for *ad*sorption onto particle surfaces.

In conclusions, Irrespective of whether the air mass is moving upward or downward, high traffic exhaust emissions and agricultural waste burning play a major role in governing the diurnal variation of carbonaceous aerosols at all sampling heights. Wind direction also appeared to make a significant difference on the atmospheric concentration of EC detected at Site-2. Daily trends also emerged, where morning OC and EC peaks appear prominently and clearly in line with the swell in traffic, whilst the afternoon reduction in mass concentrations can be attributed to the dilution effect triggered by the daytime expansion of air mass. The strong positive correlation between OC and EC observed at all sampling sites in Chiang-Mai suggests single dominant source during the sampling period.

Diurnal variation of carbonaceous aerosols reveals the highest value of OC and EC during the sampling period of 2100-0000 in Bangkok. This can be explained by i) anthropogenic activities at night market, ii) the reduction of mixing layer, iii) the decreased wind speed around midnight and iv) the termination of photolysis at night-time. The significant decrease of OC/EC ratios at Site-2 and Site-3 may be due to the stronger photolysis activities at higher altitudes coupled with the influence of aged particles from long-range transportation. The average SOC of all sampling sites was 23.5±12.3%, indicating that PM_{10} in Bangkok is considerably influenced by aged particles. The incremental lifetime particulate matter exposure (*ILPE*) model highlights the risk of carbonaceous accumulation over outdoor workers and residents living adjacent to the monitoring sites. An important day-to-night difference in the magnitude of tourism activities might have explained the large discrepancy observed between daytime and nighttime of carbonaceous composition levels in PM_{10} samples during the monitoring period. Despite of its moderate level of OC and EC contents, Hat-Yai PM_{10} contains relatively high OC/EC ratios indicating the strong influence of either aged particles derived from long-range transportation and/or biomass and agricultural waste burning aerosols. While traffic emissions play a major role in governing carbonaceous contents at ground level, wind speed and emission source type significantly influence OC and EC levels at higher altitudes.

Given the increasing number of vehicles, industrial factories, domestic hot spots and trans-boundary biomass burnings from neighbor countries, more studies are required to comprehend the carcinogenic emission source strengths and released chemical compositions from various emission sources. In the light of complicated mixing sources in Thailand atmosphere, the great challenge for government is to find the most appropriate way of reducing carcinogenic pollutants. Since PAHs are continuously released into the atmosphere, raising concerns over the safety of urban residents and those who are living or working adjacent to the emission sources, it is therefore crucial to investigate the occupation exposure to PM_{10}, carbonaceous aerosols and the dominant influence of OC/EC over the gas-particle partitioning of PAHs released from different cities. This study found the substantial risk of lung cancer in workers and residents adjacent in Pratunam (Water Gate) area, Bangkok. The analysis of $f_{SC}K_{SA}\delta_{OCT}/f_{OM}K_{OA}$ ratios indicate the overwhelming mechanisms of *ad*sorption in gas-particle partitioning of Ac, Ace, Fl, Ph, An, Pyr, Fluo, Chry and D[*a,h*]A, whereas both *ab*sorption and *ad*sorption tend to dominate the gas-particle partitioning of B[*b*]F, B[*a*]P, Ind and B[*g,h,i*]P in urban residential zone of Thailand. Interestingly, the *ab*sorption mode alone plays a minor role in gas-particle partitioning of PAHs in Chiang-Mai, Bangkok and Hat-Yai.

Acknowledgements

This work was performed with the approval of National Research Council of Thailand (NRCT). Also with acknowledged support from Institute of Earth Environment, Chinese Academy of Sciences (IEECAS) and National Institute of Development Administration (NIDA). The authors acknowledge Dr. Surat Bualert and Ms. Teeta Intasaen for their contributions on filed sampling and laboratory works.

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