Characteristics of PAHs in Particulates in the Atmospheric Environment of Hat Yai City, Thailand, and Relationship with Rubber-wood Burning in Rubber Sheet Production

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Abstract

Natural rubber is a main commodity of Thailand, half of which is formed into ribbed smoked sheets (RSS). Fuel wood is burned during RSS production to heat and dry the rubber sheets. Smoke from the burning wood contains chemical species in particles and gases, notably polycyclic aromatic hydrocarbons (PAHs). In this study, the influence of smoke from fuel-wood burning by rubber-smoking cooperatives on atmospheric air in the neighboring town of Hat Yai, Songkhla Province in southern Thailand, was evaluated for the year 2005. The particle size distributions of ambient air in downtown Hat Yai and Prince of Songkla University (PSU) clearly showed bi-modal behavior, indicating multiple sources of particles. TSP was generally low, averaging 45.6 and 46.2 $\mu g/m^3$ for downtown Hat Yai and PSU, respectively. The TSP depended on precipitation, particularly when precipitation was high. The correlation between TSP and rubber sheet production from all rubber cooperatives in Songkhla Province is clear. PAH size distributions of ambient air particles clearly showed single-modal behavior, suggesting that most of the PAHs are associated with small particles. The relationship between wood burning and PAHs in ambient air in Hat Yai and PSU is also clear. Low PAH concentrations during the dry season could result partly from wind direction (E to NE), which blows inland from the Gulf of Thailand. During the rainy season, PAH concentration at both locations was low; however, it was inversely proportional to the precipitation. Hence, rubber sheet production, wind direction and precipitation are important determinants of PAH concentration in Hat Yai area.

Keywords: PAHs, Size distribution, Wood burning, Rubber-wood, Rubber sheet

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INTRODUCTION

Thailand is the world leader in natural rubber (*Hevea brasiliensis*) production. Total rubber production in 2004 for Thailand was 2.9 million metric tons (Thailand Rubber Research Institute, 2005). Among this, 43% was formed into ribbed smoked sheets (RSS) and 80% of that was exported. China, Japan and United States are the three largest natural rubber consuming countries.

In the RSS production, fuel wood (usually rubber-wood) is burned to supply heat (and smoke) to rubber sheets in the rubber smoke rooms. Smoke from the wood burning contains fine particles and other gas species. Incomplete combustion results in the formation of polycyclic aromatic hydrocarbons (PAHs) (Furuuchi *et al*., 2006) and other chemical components. PAHs include hundreds of compounds that are carcinogenic, especially those containing 4-6 aromatic rings.

Two main problems arise from the PAHs generated from the wood burning; workplace pollution affected by the leakage of smoke particles into the working area, and atmospheric pollution to nearby surroundings. Factory workers exposed to PAHs may be at risk of developing cancer and other adverse health conditions (IARC, 1982; Kogevinas *et al*., 1998; Fracasso *et al*., 1999; Straif *et al*., 1999; Galka *et al*., 2004; Parent *et al*., 2005). Most PAHs are associated with fine airborne particles, typically 0.5 micron or less (DeMartins *et al*., 2002). These particles can travel over long ranges and may cause health problems for humans in areas surrounding the emission sources.

PAH emission characteristics from burning of various kinds of wood were investigated by Venkataraman *et al*. (2002), Hedberg *et al*. (2002), Hays *et al*. (2003). Furuuchi *et al*. (2006) studied the characteristics of smoke particles from rubber-wood combustion and evaluated the influences on workplace environments and the surrounding atmosphere. The PAH concentration inside the rubber smoking workplace was found to be extremely high, particularly of those with a larger number of aromatic rings in the fine fraction of particles. PAH concentration was about ten times higher than ambient conditions which could lead to serious health problems of the workers.

Since PAHs in particulate phase are airborne, transport of these compounds affects the quality of air in general. PAHs in atmospheric air has then become a topic of interest throughout the world. Many researchers have tried to quantify amounts of PAHs present in atmospheric air in many cities, as well as traced the sources of these compounds (Khalili *et al*., 1995; Bi *et al*., 2002; Chetwittayachan *et al*., 2002; De Martinis *et al*., 2002; Miguel *et al*., 2004; Duan *et al*., 2005).

Effects of PAHs from biofuel burning on atmospheric air were obvious. This work is then aimed at the characterization of the PAHs in the particulate phase in the atmospheric air in the city of Hat Yai, Songkhla Province in southern Thailand (Fig. 1) and how they are related to rubber-wood burning in RSS production. The city is situated in the middle of an area of factories using fuel wood, including several RSS rubber cooperatives.

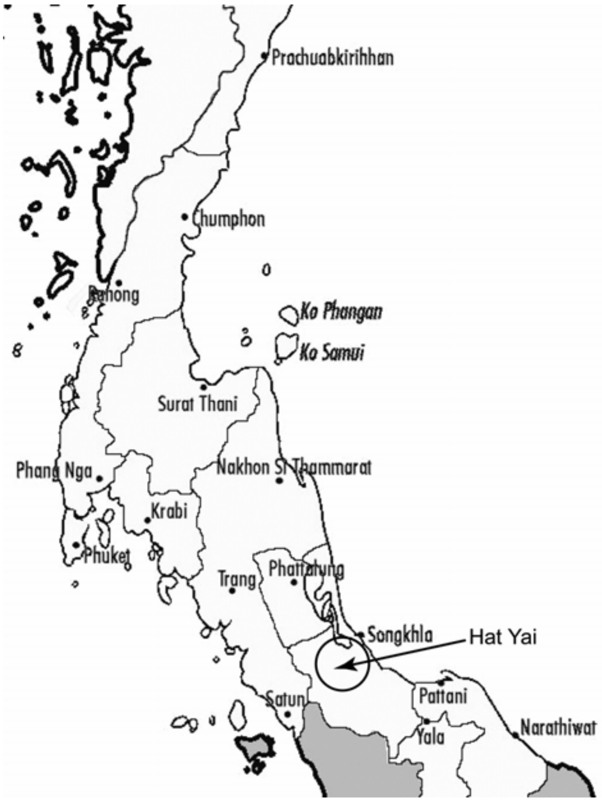

Fig. 1. Map of southern Thailand indicating the city of Hat Yai where samples were collected.

FUEL WOOD, RUBBER PRODUCTION AND WEATHER INFORMATION

The fuel wood used in RSS production is common rubber-wood abundant especially in southern Thailand. The natural rubber trees are usually cut after 25-30 years when the rubber latex productivity has reached its low point (The Thai Rubber Association, 2004). The wood used for combustion is usually in fresh condition. It is often wet from precipitation, because it is usually stored outdoors. Combustion of fresh, wet wood results in thick smoke. Total concentration of smoke particles has been found to increase exponentially by increasing the moisture content of the wood used as fuel (Kalasee *et al*., 2003). Moreover, particle concentration can be dependent on the combustion process and burning time, as well as other factors.

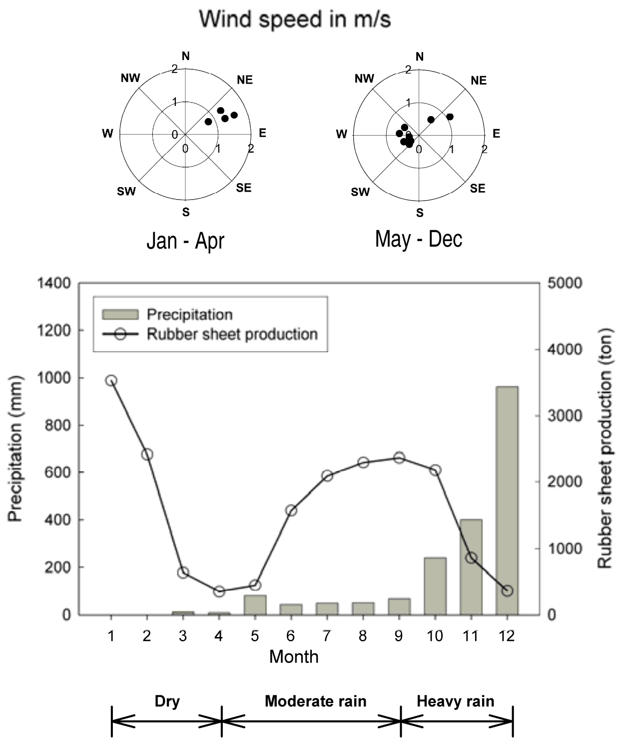

Fig. 2. Precipitation, rubber sheet productivity, and wind direction in Songkhla province.

Rubber sheet production and precipitation statistics in Songkhla Province are shown in Fig. 2. Rubber production depends strongly on the weather. In general, increased precipitation reduces rubber sheet production. The reason is that during the rainy season, farmers cannot tap the rubber latex from the rubber trees because its quality significantly drops when the latex is contaminated by the rain. Small bubbles will be present throughout the contaminated rubber sheets when dried. However, during the summer season, March to May in Thailand, the production is at its low point. This is because the rubber leaves fall during this season and latex production by the rubber trees is minimal. This explains why the rubber sheet production during March to May is very low even though there is virtually no precipitation. Rubber sheet production in 2005 peaked during January, when fuel wood usage was linearly proportional to rubber sheet production. In general, the rate of fuel wood usage was about 800-1,200 kg of fresh wood per kg of dry rubber sheets (Promtong and Tekasakul, 2007).

The weather in Songkhla Province is a typical east-coast Thailand weather pattern. During January to April, the wind direction is from east-northeast while the wind direction during May to October is from west-southwest. During November to December when precipitation is highest, the wind direction shifts back to the northeast direction. Monthly-average wind speed and direction are plotted in Fig. 2. The wind pattern affects the transport of aerosol particle in atmospheric air.

EXPERIMENTAL PROCEDURE

In this study, the influence of smoke from fuel wood burning by community-level rubber smoking cooperatives on atmospheric air is evaluated. There are about 700 such cooperatives throughout Thailand. Most of them are located in the south. Their main product is RSS. Air samplings were conducted at downtown Hat Yai and Prince of Songkla University (PSU), both in Songkhla Province next to the Malaysia border, in the year 2005. Downtown Hat Yai is a congested, heavy-traffic area, while PSU is in the suburbs of Hat Yai where the influence of traffic is low. Both locations are surrounded by several rubber smoking cooperatives.

Size fractionated particles at both locations were measured simultaneously using two 8-stage cascade (Andersen) impactors (Tokyo Dylec, AN-200). The cut-off diameters of the impaction stages are 0.43, 0.65, 1.1, 2.1, 3.3, 4.7, 7.0, and 11.0 micron. 80-mm-diameter quartz fiber filters (Advantec, QR-100) were used for smoke particle collection and for subsequent PAH analysis. They were pre-treated in a controlled environment (room temperature, 50% RH) for 72 hours. Sampling flow rate was 28.3 L/min and sampling time was 3 weeks for each sample to ensure sufficient amount of particles collected for PAH analysis. A total number of 5 samples were collected at the downtown Hat Yai station from March to September, and 2 samples were collected at PSU in March and September. Note that only one sample per month was conducted. Total suspended particulates (TSP) were measured by the commercial portable high-volume sampler (Shibata, HV500F). Sampling flow rate was set at a constant 500 L/min and sampling time was 24 hours. The filters used were identical-type quartz fiber filters (Advantec, QR-100) with 110-mm-diameters. High-volume samplings were conducted for 8 months from

March to December at downtown Hat Yai, and 5 months from February to December at PSU. One to four samples per month were collected. Only one sample in each month was analyzed for PAHs. Both the Andersen and high-volume samplers were placed at the open-roof top floors of four-story (PSU) and three-story (downtown) buildings.

All collected filter samples were then treated in the identical environment as the pre-treatment for 72 hours and then analyzed to obtain the amount of 15 different PAH components, i.e., Naphthalene (Nap), Acenaphthene (Ace), Phenanthrene (Phe), Anthracene (Ant), Fluorene (Fle), Fluoranthene (Flu), Pyrene (Pyr), Benz[a]anthracene (BaA), Chrysene (Chr), Benzo[a]pyrene (BaP), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Dibenz[a,h]anthracene (DbA), Indeno[1,2,3-cd]pyrene (IDP) and Benzo[ghi]perylene (BghiPe). A piece of each filter sample containing 4-5 mg of particulates was cut into small pieces. PAHs were extracted ultrasonically twice using ethanol:benzene (1:3, v/v) for 15 minutes. Then the solution was filtered and the filtrate was evaporated in a rotary evaporator to remove ethanol and benzene. The residue was re-dissolved in acetonitrile and kept in a refrigerator (-20 °C) for analysis. The analysis was conducted using an HPLC (Hitachi, L-2130, 2200, 2300, 2485) with a fluorescence detector along with an Inertsil ODS-P column (5 μm, 4.6 mm diameter, 250 mm length). The mobile phase was a mixture of acetonitrile and water with gradient elution from 55% acetonitrile to 100% acetonitrile at 1 mL/min and 20°C. The recovery efficiency was confirmed to be 0.82 ± 0.12 (n = 3) by spiking known amounts of external PAH standards (Accustandard, 0.2 mg/mL of each 15 PAH in 1:1 of CH_2Cl_2:MeOH) onto the pre-treated filters and then processing by the same experimental procedure used for the samples. Travel blank values of PAHs were subtracted from analyzed values. The method used in this study was modified from Toriba *et al*. (2003), Tang *et al.* (2005) and Furuuchi *et al*. (2007).

SIZE DISTRIBUTIONS AND CONCENTRATIONS

Source particle characteristics were reviewed and ambient particle size distributions and TSP, as well as PAH size distribution and concentration, were evaluated for sampled aerosol particles from downtown Hat Yai and PSU. These are discussed as follows.

Source Particle Characteristics

Kalasee *et al*. (2003) investigated the size distribution of smoke particles from rubber-wood burning using an 8-stage cascade (Andersen) impactor and found that the particle size distribution is a single (accumulation) mode. The average mass median aerodynamic diameter (MMAD) of smoke particles was found to be 0.95 micron, while the average geometric standard deviation was 2.51. Particle mass concentration depended strongly on the moisture content of the wood, ranging from 47 to 1,358 mg/m^3 for the wood moisture content from 34.5 to 107.5 % dry basis. The values of moisture content greater than 100% indicate that the weight of water in the wood is greater than that of the wood. This can occur during the rainy season where the wood is usually placed outdoors and is exposed to rain for long periods.

Furuuchi *et al*. (2006) found that the total PAH concentration of particles from the rubber-wood combustion source is very high. The PAH concentration for particles smaller than 3.3 micron is higher than 10^5 ng/m^3, while the concentration for larger particles is about 10^3 ng/m^3. It was found that the fractions of Phenanthrene (Phe), Pyrene (Pyr), and Fluoranthene (Flu), have the highest PAH compositions from source particles; and fractions of PAHs with larger molecular sizes (4-6 rings) are also high, probably because these compositions are usually generated from biomass combustion.

Ambient Particle Size Distribution and TSP

The particle mass size distributions of ambient air in downtown Hat Yai sampled from March to September, 2005 are shown in Fig. 3. It clearly displays bi-modal behavior where the accumulation-mode peaks take place at the particle size of 0.54 micron, while the coarse-mode peaks take place at 4.0 micron (see definitions of atmospheric aerosol modes in Friedlander, 2000). Concentration of accumulation-mode particles is comparable to that of the coarse-mode particles, though the size is smaller. Sources of accumulation-mode particles could be from either biomass burning (Mukherji *et al*., 2002), or diesel particulate matters (Vogt *et al*., 2003). Fuel wood, especially rubber-wood, is the most common and cheapest fuel used in industries around the city, including rubber smoking cooperatives and factories, and in the households outside of the town. Note that diesel-engine automobile is one of the dominant modes of transport in Hat Yai, as well as many other cities in Thailand. Most of the diesel-engine vehicles are old and emit black smoke, which is likely to contribute to the particles in the accumulation mode in the downtown area. The sources of coarse-mode particles could be from re-suspension of ground dust or other atmospheric aerosol particles.

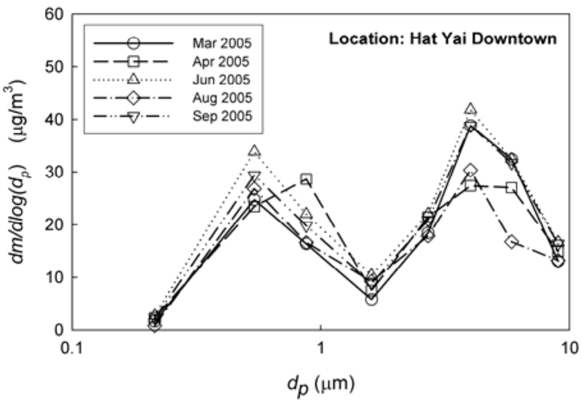

Fig. 3. Size distribution of particulate matters in Hat Yai downtown.

Particle size distribution at PSU also shows two modes as in the case of the downtown, as shown in Fig. 4. The accumulation-mode particles are likely from biomass burning, since the traffic around PSU is light and contribution from diesel engines should not be significant.

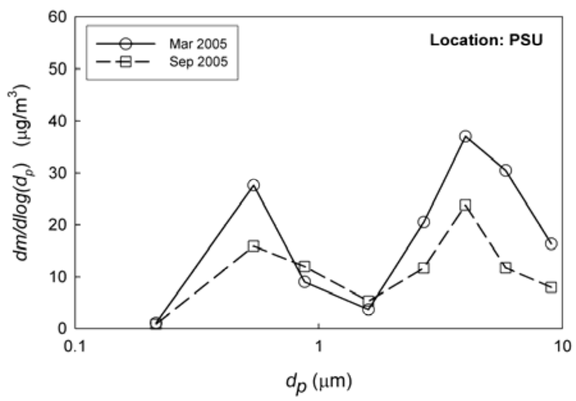

Fig. 4. Size distribution of particulate matters in PSU.

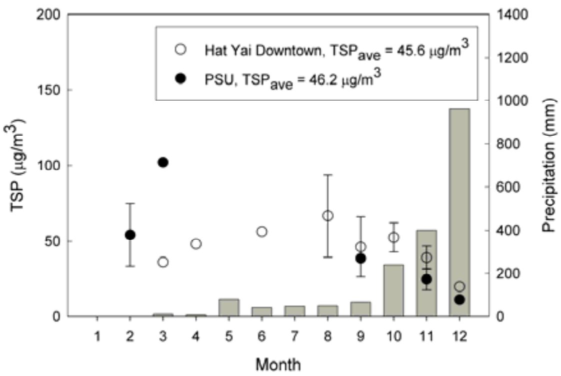

Fig. 5. TSP and precipitation in 2005.

The total suspended particulates (TSP) of air sampled from downtown Hat Yai and PSU by high-volume sampler are shown in Fig. 5 along with average precipitation in Songkhla Province. The yearly average TSP is 45.6 and 46.2 μg/m^3 for downtown Hat Yai and PSU, respectively. The values are almost identical and rather low compared with standards (330 μg/m^3) regulated by the Thailand Pollution Control Department (National Environment Board, 1995). Relationship between TSP and precipitation is shown in Fig. 6. It is obvious that TSP depends on precipitation, particularly when precipitation is high. TSP is decreased almost linearly when precipitation is increased, though there is a small period when the TSP is increased with increasing precipitation. This takes place when the precipitation is lower than 200 mm/month, the dry season (March to June). This could be an effect of long-range summer forest-fire aerosol particle transport from our neighboring countries, especially Indonesia. However, the effect is neither strong nor obvious. The correlation between TSP and rubber sheet production from all rubber cooperatives in Songkhla Province shows that increase of rubber sheet production

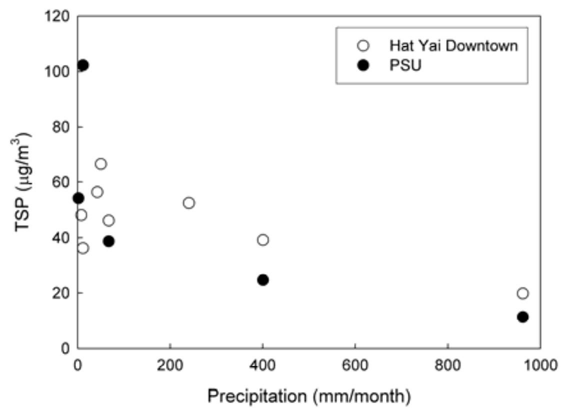

Fig. 6. Relationship between TSP and precipitation.

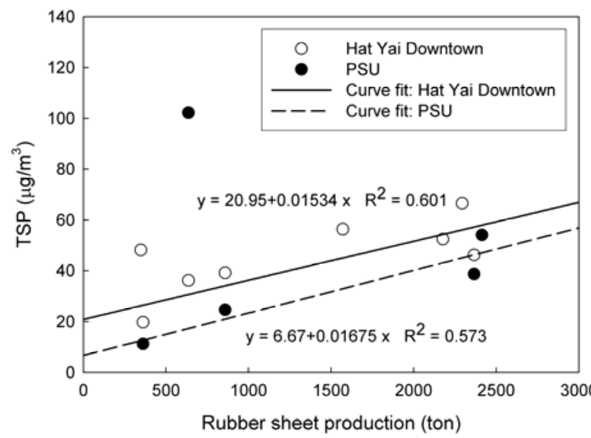

Fig. 7. Relationship between TSP and rubber sheet production.

almost linearly enhances the TSP both at downtown Hat Yai and PSU, as shown in Fig. 7. Hence the contribution of smoke particles generated from rubber-wood burning to TSP is quite clear. Source apportionment of the particles will be discussed next.

Ambient PAH Size Distribution and Concentration

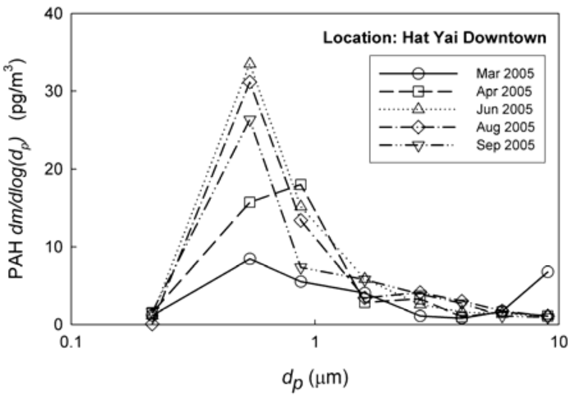

Fig. 8. Size distribution of total PAHs on particulate matters.

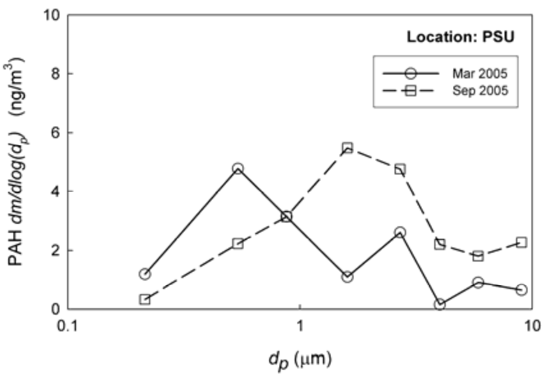

Fig. 9. Size distribution of PAH concentration.

PAH mass size distributions from Andersen samplings at both locations, analyzed by an HPLC, are shown in Figs. 8 and 9, respectively. They are mostly single mode. It can be concluded that most of the PAHs are associated with small particles. At Hat Yai downtown, the peak takes place at the average aerodynamic diameter of 0.54 micron, while the peaks at PSU take place at 0.54 and 1.6 microns for March and September, respectively. The PAH size distribution at downtown Hat Yai is in accordance with the size fraction of the aerosol which shows a high concentration at the same diameter (0.54 micron). However, at PSU, the peak for PAH distribution sampled in September takes place at the slightly larger particle size than the particle size distribution.

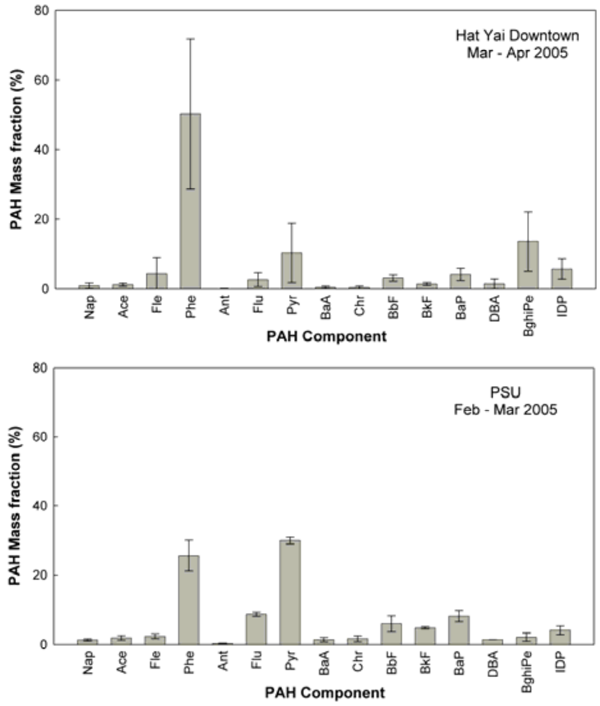

Fig. 10. Mass fraction of PAH compounds from high-volume sampling in dry season in (a) Hat Yai downtown, and (b) PSU.

Mass fractions of PAHs from both downtown Hat Yai and PSU in the summer (dry) season (January to April) are shown in Figs. 10 (a) and (b), respectively. Both locations show effects of PAHs from wood burning as Phenanthrene (Phe), Pyrene (Pyr), and Fluoranthene (Flu) are high, particularly in PSU. However, the presence of large molecular PAHs (5-6 rings) is not obvious. Effects of wood burning PAHs is more obvious in PSU may be due to the location of PSU which is close to several rubber cooperatives and other wood-burning sources. Direction of wind may play some role in this behavior of PAHs as in the dry season wind blows from the Gulf of Thailand to the town (northeast wind) as shown in Fig. 11. Since PSU is situated west from a mountain, the effect of the wind on PSU sampling may not be as significant as that on Hat Yai downtown.

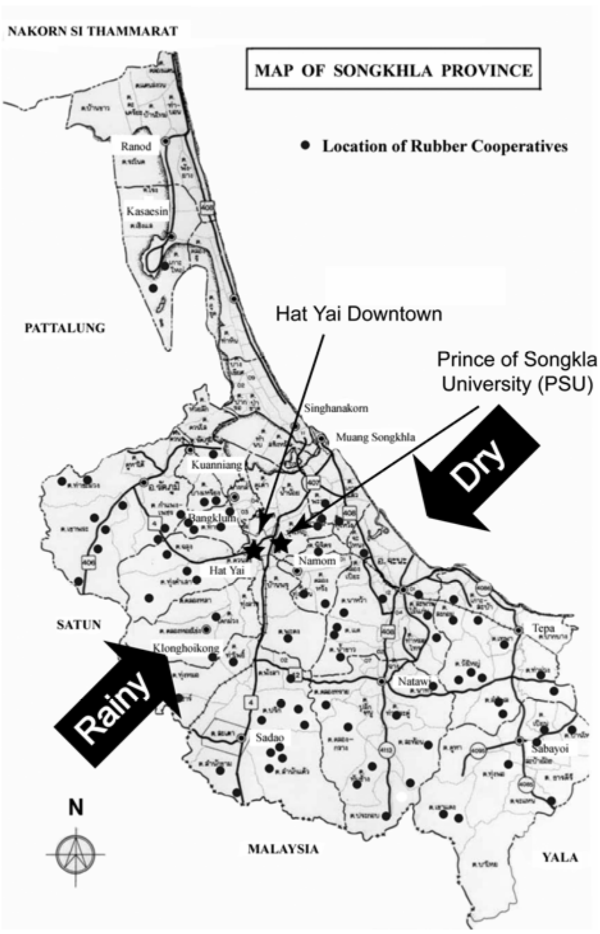

Fig. 11. Wind direction in Songkhla Province during dry and rainy seasons.

PAH profiles with large fractions of biomass origin composition were detected in downtown May-November as shown in Fig. 12 (a), and PSU September-December in Fig. 12 (b). Precipitation during these periods was moderate to high. Results from both locations show similarity in PAH characteristics. This indicates the possibility of the transportation of smoke particles from cooperatives as well as other wood burning sources to downtown Hat Yai and PSU.

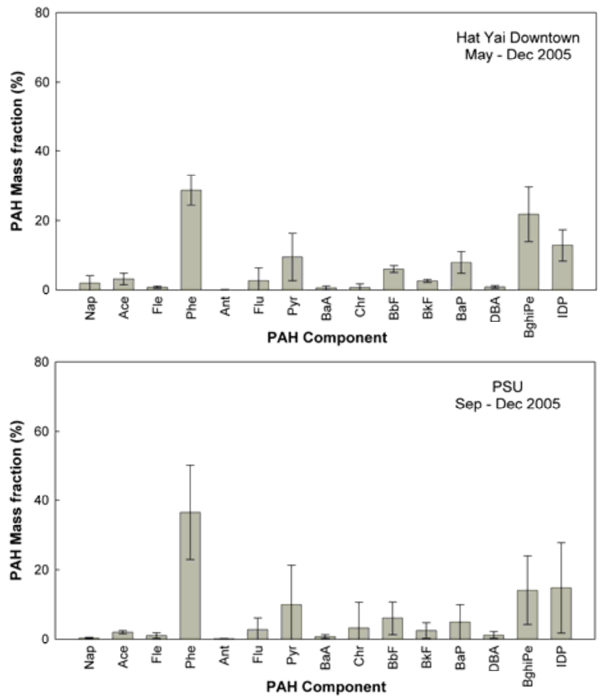

Fig. 12. Mass fraction of PAH compounds from high-volume sampling in rainy season in (a) Hat Yai downtown, and (b) PSU.

Fig. 13 shows the PAH concentration profiles from both downtown Hat Yai and PSU along with the rubber sheet production in Songkhla Province. PAH concentration is defined as the summation of 15 components analyzed. During the dry season (January –

April), PAH concentrations are low in both locations. In this season, production of rubber sheet is high in January and starts to decrease in February due to the weather. Rubber sheet production determines the amount of fuel wood used in drying the sheets which proportionally indicates the amount of the smoke generated. In March and April, which are the peak months of summer season in Thailand, the rubber sheet production reaches the lowest point as this is the season when leaves of the rubber trees fall and their latex production is lowest. The low concentration of PAH could partly result from the wind direction (E to NE) as mentioned earlier. From May to December, PAH concentration is directly related to rubber sheet production.

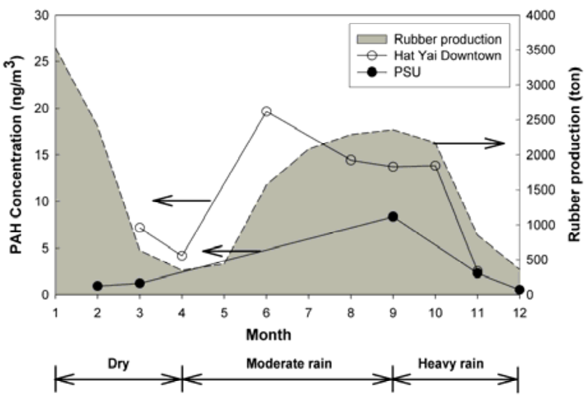

Fig. 13. Rubber sheet production and PAH concentration during samplings in 2005.

The relationships between 4-6 rings PAHs and rubber sheet production are shown in Figs. 14 (a) and (b), for PAH concentration and mass fraction, respectively. The correlations exhibit exponential function of PAH concentration (or mass fraction) with respect to rubber-sheet production. All correlations are good with r-squared values exceeding 0.8 (Tables 1 and 2). It is clear that the PAH concentration or mass fraction at both locations are nearly the same during the rainy season, but they vary considerably in the dry season. This could be the result of geography and wind effects. During the rainy season, the wind is usually blowing southwesterly towards the ocean. Both locations should be exposed to the same sources of emission at this time. In contrast, during the dry season, the wind blows from the ocean (NE). PSU, located at the foot of the mountain downwind, may receive less effect of the PAHs from

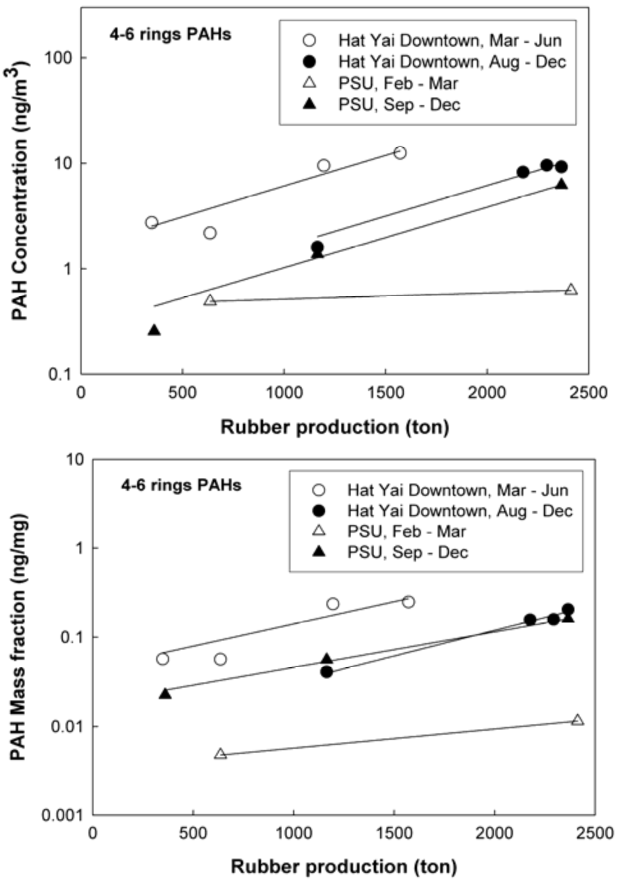

Fig. 14. Relationship between the amount of 4-6 rings PAH and rubber sheet production (a) PAH concentration, (b) PAH mass fraction.

Table 1. Parameters for the fitted equations for PAH (4-6 rings) concentration using relation $y = a e^{bx}$.

Parameters	HY DT Mar-Jun	HY DT Aug-Nov	PSU Feb-Mar	PSU Sep-Dec
a	1.587	0.4307	0.4497	0.2739
b	0.001343	0.001329	0.000135	0.0013200
r^2	0.9324	0.9715	1.0	0.9977

Table 2. Parameters for the fitted equations for PAH (4-6 rings) mass fraction using relation $y = a e^{bx}$.

Parameters	HY DT Mar-Jun	HY DT Aug-Nov	PSU Feb-Mar	PSU Sep-Dec
a	0.04479	0.008538	0.003505	0.01843
b	0.001148	0.0013200	0.000492	0.000916
r^2	0.8396	0.9671	1.0	0.9985

upstream sources than Hat Yai's downtown sampling station about 3 km away.

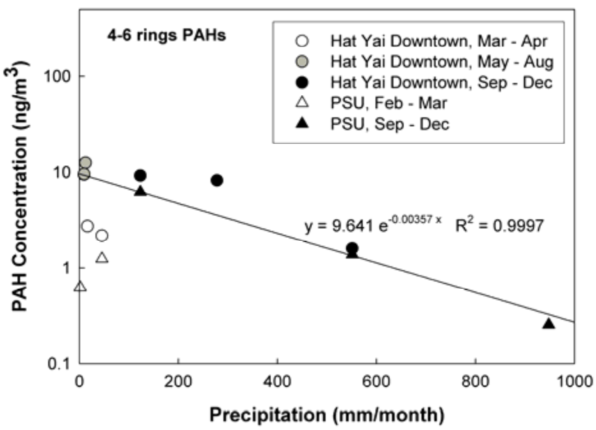

Fig. 15. Relationship between PAH concentration and precipitation.

Fig. 15 shows the relationship of the PAH concentration and precipitation. During the dry season (January–April), the PAH concentrations at both locations are low as mentioned and correlations with precipitation are not clear. During the moderate-rain (May–August) and heavy-rain periods (September–December), however, PAH concentration is exponentially decreased when the precipitation increases. Hence in these periods, rubber sheet production, wind direction and precipitation are important to PAH concentration in the Hat Yai area.

CONCLUSION

The particle size distributions of ambient air in downtown Hat Yai and PSU clearly show bi-modal behavior where the accumulation-mode peak takes place at the particle size of 0.54 micron, while the coarse-mode peak takes place at 4.0 microns. Total suspended particulates of air sampled from both locations are generally low, averaging 45.6 and 46.2 μg/m^3 for downtown Hat Yai and PSU, respectively. It is quite obvious that the TSP depends on precipitation, particularly when precipitation is high. However, in the dry season, TSP does not show clear dependence on the precipitation at either location. The correlation between TSP and rubber-sheet production from all rubber cooperatives in Songkhla Province is clear.

PAH size distributions of ambient air particles in both locations clearly show single-modal behavior. It can be concluded that most of the PAHs are associated with small particles. Effects of wood burning on ambient air in downtown Hat Yai and PSU are clear although it is more obvious in the rainy season than in the dry season. This is probably due to the wind directions. The majority of wood-smoke generation sources are located upwind in the rainy season. The correlation between PAH concentration and rubber-wood burning is also clear. The low concentration of PAH in the dry season could partly result from the wind direction (E to NE) which blows inland from the Gulf of Thailand. During the rainy season, PAH concentration is inversely proportional to the precipitation. Hence in these periods, all three factors — rubber sheet production, wind direction and precipitation — are important to PAH concentration in the Hat Yai area.

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