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Ambient and Indoor PM10 and PM10-bound PAHs around the Vicinity of an Industrial Estate in Rayong, Thailand: Concentration and Source Identification

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Abstract

The ambient and indoor air concentrations of PM₁₀ and PM₁₀-bound PAHs (16 priority PAHs) were investigated in Rayong Province, Thailand. The locations of the selected study areas were Map Ta Phut Industrial Estate and its vicinity comprising six sampling sites (four industrial areas and two vicinity areas). The indoor and ambient air samples were collected in March 2017. The sampling sites located close to the road were detected the greatest ambient average concentrations of PM₁₀, with an average value of 56.06 μ g m⁻³ (Map Chalut Area or MC). The levels of indoor PM_{10} were mostly dependent on the resident activities and the highest mean level of PM_{10} was 31.29μ g m⁻³ detected at Huai Pong. The Benzo(b)fluoranthene and acenaphthylene were the major PAHs found to have the highest 24-h average concentrations for both indoor and ambient air. The highest mean ambient and indoor air levels of benzo(b)fluoranthene were 49.18 and 30.88 ng m⁻³, respectively, found at MC. In terms of 16 total PAHs, MC was found to have the greatest level. Analysis of the diagnostic ratios determined that the traffic density was the major source of influence on particle-bound PAH concentrations for both ambient and indoor air samples, which indicated that the greater the traffic volume, the higher the level of PAHs.

Keywords: PM10; PAHs; Thailand; Source identification; Industrial area

Introduction

Map Ta Phut, the largest industrial site of Rayong Province, Thailand, consist of petrochemical companies, oil refineries, heavy industries, steel companies, chemical fertilizer companies and private industries [1]. Environmental problems are the biggest issues of this area, especially soil and air pollutions due to human activities such as oil spill, shipping emissions, industrial processes and etc. [2]. The previous research has revealed that particulate matters and the persistent organic pollutants (Polycyclic Aromatic Hydrocarbon or PAHs) were the crucial sources of air pollutants in industrial areas [3]. These toxic pollutants (particulate matter and PAHs) are suspected to cause health effects in the employees as well as the residents who live around this industrial area. In addition, the health problems of these people are expected to increase the chances of respiratory diseases and cancer [4]. Thus, concerned for air pollutants in this area due to the industrial processes along with the transportation sectors is warranted.

Particle pollution is a dominant cause of many health effects. There are many health problems from exposure to particulate matter with a diameter of less than 10 μ m (PM₁₀). Exposure to PM_{10} can affect the heart and lungs, causing chronic bronchitis and emphysema [5–6]. Also, other pollutants that bound to PM_{10} , such as PAHs, should be a concern. PAHs are generated from both anthropogenic and natural emissions. For anthropogenic sources, PAHs are mainly formed from the incomplete combustion of vehicles and industrial processes (such as coal, oil, petrol and biomass) [7–12], besides the fireworks are also concerned as the main source of PM10-bound PAHs [13]. In the case of natural sources, PAHs are produced from volcanic activities and open burning [14]. The structures of PAHs are comprised of two to seven aromatic rings, and the low molecular weight PAHs (LMW PAHs) with high vapor pressure tend to occur in the gas phase. On the other hand, high molecular weight (HMW PAHs) with low vapor pressure PAHs tend to bond with particles [15]. The United States Environmental Protection Agency, or U.S. EPA., classified PAHs as a hazardous air pollutant which is suspected to cause cancer [16]. The 16 PAHs including Benzo

(a)pyrene (BAP), Dibenz(a,h)anthracene (DA), Benzo(a)anthracene (BAA), Benzo(b)fluoranthene (BBF), Benzo(k) fluoranthene (BKA), Chrysene (CS), Naphthalene (NT), Indeno(1,2,3cd)pyrene (IP), Acenaphthene (AC), Anthracene (AN), Acenaphthylene (AT) , Benzo (g,h,i) perylene (BGP), Fluoranthene (FT), Fluorene (FR), Phenanthrene (PT) and Pyrene (PR) have been recognized as the High Priority Pollutants by the U.S. EPA due to the potential toxicity, occurrence and persistence in the environment. A group of sixteen PAHs, particularly benzo(a) pyrene, are found to be the most toxic.

Normally, the level of PAHs in the industrial area is greater than those observed in the nonindustrial and rural area due to the industrial activities [17]. Correspondingly, the associated factors influencing the PAH profiles were the number of vehicles and traffic volume. Furthermore, the vicinity of industrial area can result in a high level of PAHs as well. These compounds are also found as the predominant air pollutants in urban areas caused by human activities. Mostly, the ambient air pollutants can penetrate into the indoor air environment via the cracks in buildings, windows and doors [18–19]. The emission sources of indoor PAHs are dependent on both outdoor air and resident activities. Normally, people spend 90% approximately [20] so indoor PAH levels should be of concern as well as the ambient PAHs because it can cause many health problems. The studies of PAHs bound to fine particles are predominant, whereas information on PAHs bound to coarse particles is lacking. Moreover, the study of PM10-bound PAH levels in Map Ta Phut Area is still negligible for both indoor and ambient environment. In light of the high PAH concentrations, the study of the residents living within the Map Ta Phut is very important for both industrial and non-industrial areas. Therefore, the industrial areas with high PAH levels were investigated in this study and compared with the PAH levels of the nonindustrial areas.

Materials and methods

1) Air sampling site

In this study, air samples were collected around Map Tha Phut, Rayong Province. The six communities of the selected study areas were located within both industrial area and its vicinity (non-industrial area). The ambient and indoor air samples were collected at six locations around Map Tha Phut area (see Figure 1 and Table 1). Both indoor and ambient air samples were monitored in the industrial area and its vicinity. Samples from four industrial areas and two vicinity areas were collected in March 2017. To assess indoor air pollution, two residential buildings were observed in each community (12 houses were collected from the 6 studied communities) to investigate both PM₁₀ and PAHs. Air samples were collected in triplicate at each location.

2) Sample preparation and analysis

Sampling of PM_{10} concentrations in the ambient air was done using PTFE filters with a diameter of 47 mm, pore size 2 μm. The Deployable particulate sampler system was operated at a flow rate of 10 L min⁻¹. For PM_{10}

Table 1 The description of sampling sites

concentration in indoor air sampling, PAHs were collected using SKC pump Model P45XE connected to cyclone and PTFE filters with a diameter of 37 mm, pore size 2 μm, and a flow rate of 2 L min⁻¹. The sampling period of both indoor and ambient air was 24 h d⁻¹ continuously for 3 d.

Figure 1 Locations of the sampling sites.

In the case of PM10-bound PAHs measurement, the PTFE sample filters were placed into amber vials and 15 mL of dichloromethane was added. After that, the solution was separated by using syringe filter. Next, 50 µL of dimethyl Sulfoxide (DMSO) was added to the solution and the dichloromethane purged by nitrogen (99.99%) with a heating box at 35ºC. The extracted solution was kept in the freezer until analysis by High Performance Liquid Chromatography (HPLC/GC) with Fluorescence and UV-VIS detector, model Shimadzu SPD 20A. This method was modified from Method5506 NIOSH Manual of Analytical (1998).

The limit of detection (LOD) and limit of quantification (LOQ) of HPLC-UV were investigated by injection of PAHs Standard (Supelco, USA) that corresponded to a signal-to-noise value that greater than or equal to 3. The standard deviation (SD) of concentration of each congener was determined, and 3SD and 10SD were defined as the LOD and LOQ, respectively. The reference values of LOD and LOQ quantification ranged from 0.0032 to 0.0183 mg L^{-1} . The recovery was performed by adding known amount of mixture standard of PAHs and then the spiked samples were then extracted and quantified. The average percent recovery for PAHs was ranged from 70 to 120.

3) Calculation of PAHs concentrations

The 16 PAHs (total PAHs) analyzed were Benzo(a)pyrene or BAP, Dibenz(a,h)anthracene or DA, Benz(a)anthracene or BAA, Benzo(b) fluoranthene or BBF, Benzo(k)fluoranthene or BKA, Chrysene or CS, Naphthalene or NT, Indeno(1,2,3cd)pyrene or IP, Acenaphthene or AC, Anthracene or AN, Acenaphthylene or AT, Benzo(g,h,i)perylene or BGP, Fluoranthene or FT, Fluorene or FR, Phenanthrene or PT and Pyrene or PR. The calculation of PAH concentration after analyzing with HPLC-UV could be determined by using the Eq. 1.

PAHs conc. =
$$
\frac{(CA - CB) \times VS \times CF}{Vair}
$$
 (Eq. 1)

where PAHs conc. is concentration of PAHs (ng m-3), CA is concentration of PAHs in each sample $(mg L^{-1})$, CB is concentration of PAHs in the blank (mg L^{-1}), CF is conversion factor $(10^6 \text{ ng mg}^{-1})$, VS is the sample volume of 100 μ L, and V_{air} is air volume (m³).

4) Data analysis

The possible sources of both ambient and indoor PAHs were formulated by using the diagnostic ratios (DRs). The DRs of AN/ (AN+ PT), BAA/(BAA+CS), IP/(IP+BGP), and FT/ (FT+PR) were analyzed in this study. SPSS for Windows version 20 software was utilized for statistical analysis in this study. The significant differences between indoor and outdoor PM10 bounded PAHs were analyzed by using T-Test. The comparison of PAH congeners among the six air quality observatory sites were analyzed using ANOVA. A p-value of 0.05 was accepted for all cases.

Results and discussion

1) Ambient concentration of PM10

Daily average ambient air concentrations of PM₁₀ at the six sampling sites are summarized in Figure 2. All detected PM10 levels did not exceed the 24-h PM₁₀ standard of Thailand (120 μ g m⁻³) [21]. The ambient PM₁₀ concentrations and standard deviations of NF, KY, MC, WS (industrial area) were 40.33±11.15, 20.30±4.32, 56.06±12.68, 34.40±10.33. For non-industrial areas, the ambient levels of HP and CP sites were 43.13 ± 20.14 , and 30.80 ± 5.86 µg m⁻³, correspondingly. MC site was found to have the greatest ambient PM10 concentration because this sampling site was located near the street and a crematory. Thus, the factors of transportation sector and the process of cremation might directly affect the PM_{10} concentration. Several types of air pollutants also originate from cremation, including coarse particles (PM_{10}) [22]. On the other hand, the lowest concentration of PM₁₀ was observed at KY caused by this sampling site being far from the side street. From the comparison of PM₁₀ levels determined in the present study with those determined in other studies, the highest PM_{10} concentrations were detected in India (196 μ g m⁻³), followed by samples taken in this study (38 µg m^{-3}) , Spain (25 μ g m⁻³ for La Coruna and 33 μ g m⁻³ for Barcelona) and Italy $(24 \mu g m^{-3})$, respectively $[23-26]$. The average levels of PM₁₀ in Thailand were about 5 times lower than those measured in Kolkata (India), but comparable with those investigated in Italy and Spain.

2) Indoor air concentration of PM10

Two households were taken in each sampling site and the results are reported in Figure 2. Since Thailand does not have regulatory standard for the indoor air quality, the results of indoor PM₁₀ in this study were compared with the indoor air quality of the World Health Organization (WHO) guideline $(50 \text{ µg m}^3 \text{ for } 24\text{-h mean})$ and China air quality standard $(50 \mu g m^3)$ for 24-h mean) [27]. All PM10 levels of indoor air recorded in this study were observed to be lower than 50 μ g m⁻³. The trend of PM₁₀ concentrations was similar in indoor sampling for all communities. The sampling houses of HP had the highest mean PM_{10} concentration (26.49 \pm 8.06 μ g m⁻³), followed by WS (21.94±22.81 μ g m⁻³), KY (19.42±9.30 μg m⁻³), NF (17.30±11.24 μg m⁻³), MC (15.11±11.15 μ g m⁻³) and CP (12.61± 6.75 μ g m⁻³), correspondingly. The greatest PM₁₀ levels were shown in HP site because the owner of these houses were the truck drivers, and the trucks were parked in front of their houses. Consequently, the detected of PM10 levels may be coming from the truck's flue gases in the exhaust pipe. Similarly, these sites also located close to the side street and the traffic density around this area was slightly high. For WS,

PM₁₀ value of WS2 was greater than that found in WS1 due to the location and activities of residents in WS2. Additionally, the residents in WS2 burnt incense every day and the location of this site was near the street. For KY community, a high level of PM_{10} was due to the activity of the residents, which included burning incenses and cooking using biomass fuels (KY1). On the other hand, the house with lower PM₁₀ level (KY2) was located far from the street, so the PM concentration was observed in low level. Considering the CP sampling site, PM₁₀ concentrations were lower than those found in other areas which can explained by the location of this sampling site being situated in a non-industrial area. Equivalently, the sampling sites of this community were situated too far away from both main road and side street.

Figure 2 PM₁₀ concentration in each community.

3) The indoor PM10-bound PAHs concentration

The indoor concentrations of PAHs in six sampling sites (12 houses) are illustrated in Figure 3 and 5. The greatest mean total PAH concentrations of indoor air were found in MC $(30.34 \pm 7.82 \text{ ng m}^{-3})$, followed by HP $(25.00 \pm 7.82 \text{ mg m}^{-3})$ 6.23 ng m⁻³), WS (15.81 \pm 4.75 ng m⁻³), CP $(14.90\pm3.97 \text{ ng m}^3)$ and KY $(10.73\pm2.31 \text{ ng m}^3)$, correspondingly. Indoor PAHs resulting from burning incense is a dominant source. The highest levels of total PAHs were determined in

MC community (43.54 for MC1 and 17.13 ng m⁻³ for MC2) because the residents of MC1 always burnt 20 to 30 incense sticks to worship Gods and ancestors. In all communities, the results showed that BBF was the major specie among all PAHs compounds followed by AT. This may be caused by the activities of the household residents which included both cooking (BBF) and incense burning (AT). The main source of BBF came from the traditional cooking stove that contained the large amounts of PAHs [28]. In view of the AT concentrations, the houses of the residents who burn incense inside exhibited the greater levels of this chemical than those of the houses of residents who do not burn incense in their own units. Some studies revealed that AT was primarily produced from burning incenses that depended on the type of incense [29]. In addition, the transportation sector emitted AT as well [30], so the sampling site of HP2 that was located next to the side street and truck parking revealed AT even though this household was not burning incense. Not only do burning incense and combustion engine vehicles generate AT, but cigarettes also induced this substance. From this evidence, the third day of sampling at CP2 which situated well away from side street and no incense burning activity still showed detectable AT level and this may be caused by cigarette smoking. As the result, one of the family members of CP2 smoked in the house on the third day sampling. Other researchers investigated that this chemical was also released from the traffic, in addition to cooking and heating [31]. The type of fuels, especially coal and biomass that used for cooking and heating were suspected to released PAHs. The evaluation of the significant differences among indoor PAH congeners detected at six air quality observatory sites were considered using ANOVA. The results showed the significantly different between BBF and the other PAH congeners for all sampling sites.

Generally, PAHs comprised of LWM PAHs (containing two to three benzene rings) and HWM PAHs (containing four or more rings). Two aromatic rings are comprised of NT; three aromatic rings including AT, AC, FR, PT and AN; four aromatic rings including FT, PR, BAA and CS; five aromatic rings including BBF, BKA and BAP; six aromatic rings including IP, DA and BGP. Mostly, HMW PAHs were come from gasoline vehicles, whereas LMW PAHs were originated from the coal combustion [32]. Comparing the indoor PAH congeners among six air quality observatory sites at Map Tha Phut by using ANOVA, there were no significant differences among six sampling sites for NT, AT, AC, FR, PT, AN, FT, PR, BAA, CS, BKA, BAP and IP. Nevertheless, BBF had the significantly different between HP and NF, MC and KY, and MC and NF communities. DA showed the significantly different between MC and KY, and MC and NF, whereas BGP had the significantly different between CP and NF, HP and NF, KY and NF, MC and NF, and WS and NF. For total PAHs, there were significantly different between HP and NF, MC and KY, and MC and NF. From overall results, there were no significant differences among six sampling sites for several PAH congeners, excepted for BBF, DA and BGP (HWM PAHs). In view of BBF, DA and BGP, significant differences were mostly investigated between MC site and the other sampling sites. Consequently, the indoor PAH levels of MC site are significantly influenced by gasoline vehicles because this site was situated next to the side street. Moreover, the activities of the household residents were affected and influenced the levels of indoor PAHs too.

Figure 3 Indoor air concentration of PAHs in (a) industrial area (b) non-industrial area.

The comparison of total PAH concentrations in this study with other studies is shown in Table 2. The levels of total PAHs in Rayong Province were lower than those of the previous study recorded in China. The PAH levels were related to human activities, including vehicle emission, biomass and coal combustion [33]. The concentrations of total PAHs in Chiang Mai were lower than those conducted in Lampang and Rayong, Thailand, as well as those reported in Menen, Belgium.

4) The ambient PM10-bound PAHs concentration

The ambient PAH levels are depicted in Figure 4 and 5. The levels of the total PAHs in MC (67.07 \pm 29.61 ng m⁻³) were greater than those detected in HP $(36.06 \pm 28.74 \text{ ng } \text{m}^{-3})$, CP $(33.68 \pm 8.13 \text{ ng m}^{-3})$, NF $(31.74 \pm 17.69 \text{ ng m}^{-3})$, KY (27.77 \pm 43.31 ng m⁻³) and WS (26.80 \pm 10.59 ng m-3), respectively. BBF was identified from traffic and industrial activities as the major source of PAHs emission. Hence, the ambient air sampling of both industrial and non-industrial areas had the highest concentrations of BBF that were predominantly promoted from the transportation. The MC community appeared to have the top-level of BBF $(49.18 \text{ ng m}^{-3})$, followed by AT $(21.85 \text{ ng m}^{-3})$ because this site was located near the side street and parking lot. Apart from these two types of chemical substances, the HMW PAHs which composed of BKA, BAP, DA, BGP and IP were detected in the ambient air. These HMW compounds, particularly BBF and DA were usually obtained from high traffic areas rather than that observed in the background areas [31]. Therefore, the concentrations of these substances in the ambient air occurred much greater than that found in the indoor air. The evaluation of the significant differences among

ambient PAH congeners detected at six air quality observatory sites were considered. The results showed the significantly different between AT and the other PAH congeners, and BBF and the other PAH congeners for all sampling sites, excepted for MC. Only MC site showed the significantly different between BBF and the other PAH congeners.

Comparing the ambient PAH congeners among six air quality observatory sites, there were insignificant difference among all sampling sites for NT, AT, AC, PT, AN, BKA, DA, BGP and total PAHs. Nevertheless, FR had the significantly different between CP and KY sites, while FT showed the significantly different between MC and KY sites. PR had the significantly different between CP and WS, and MC and WS. For CS, there were significantly different between CP and MC, MC and HP, MC and WS, MC and KY, and MC and NF. From the statistical analysis data, significant differences were mostly observed between MC site and the other sampling sites, particularly HMW PAHs (FT, CS, BBF, BAP and IP). Therefore, it could be found that MC site which located next to the side street, indicating PAHs in the atmosphere of this site that are significantly influenced by gasoline vehicles.

Sites	Features	Average concentration $(ng m-3)$	References
Rayong, Thailand	Industrial area	16.2	This study
	Vicinity area of		
	industrial estate	19.9	This study
Lampang, Thailand	Rural area	$4.2 - 224$	$[34]$
Chiang Mai, Thailand	Urban area	6.7	$\left[35\right]$
Menen, Belgium	Industrial area	6.6	$[36]$
China	Rural area	770 (heating seasons)	$[37]$
		312 (non-heating seasons)	
	Urban area	690 (heating seasons)	
		404 (non-heating seasons)	
	Urban area	209	$\lceil 33 \rceil$
	Rural area	199	

Table 2 Average indoor concentrations of PAHs in other locations

Figure 4 Ambient concentrations of PAHs at all communities.

Figure 5 Average concentrations of total PAHs.

For comparison of the outdoor and indoor PAH levels, the significant differences between indoor and outdoor PM10-bounded PAHs were analyzed by using T-Test. For HP and KY sites, there were no significant differences in the 16 PAHs between indoor and ambient PAH levels. For NF community, the BAA, BBF, DA and IP levels had the significantly differences between indoor and outdoor samples. In view of CP community, the levels of indoor NT, AT, FR, FT, PR, BAA, BKA, BAP, DA and IP were significantly different from the ambient air. In addition, the NT and BAP levels showed the significant differences between the indoor and outdoor PM10-bounded PAHs in MC site. The concentrations of LMW PAHs (AT and FR) revealed the significant differences between the ambient and indoor levels in WS community. From overall results, the PAH levels of CP and NF sites in the ambient air were significantly higher than those in the indoor air, principally for HMW PAHs. Accordingly, the atmospheric transport might play a vital role for the ambient PAH levels at CP and NF sites when compared with the indoor PAHs. Nonetheless, no significant differences were found for total PAHs between indoor and outdoor for all communities.

5) Possible sources of PAHs

The diagnostic ratios of PAHs were applied to classify and categorize pollution emission sources for ambient and indoor environment. Additionally, it is a simple and convenient approach for classification of atmospheric PAHs in order to provide a broad comprehensive perspective of source identification [38]. Anyhow, the chemical reaction with other air pollutants should be concerned, especially oxidizing gaseous species from vehicular exhaust. The heterogeneous reactions between oxidizing gaseous species (ozone, $NO₃/N₂O₅$ and OH radicals) from vehicular exhaust and LMW PAHs were mainly intended to reduce the concentrations of AN and PT [38]. Correspondingly, the source categorization of PAHs by using the binary ratios of AN/(AN+PT) might affect the alteration of AN/(AN+PT) ratios, especially the high traffic areas. Although the locations of all sampling sites in this study were situated close to the side street, they were not situated in the areas of heavy traffic congestion areas. For this reason,

the reduction of LWM PAHs, causing the alteration of AN/(AN+PT) ratios from the heterogeneous reactions was play a minor role in this study. The ratios of indoor FT/(FT+PR), AN/(AN+PT), BAA/(BAA+CS) and IP/(IP+ BGP) are demonstrated in Figure 6. The results proved that the dominant sources of PAHs in the industrial and non-industrial areas were tended to be emitted from the traffic. These kinds of PAHs were transported and penetrated into the house (indoor air). Identically, IP/(IP+ BGP) ratio of some sampling sites demonstrated that solid combustion and wood burning were the important sources of PAHs and it is consistent with the fact that there was incense burning in the actual household environment. Therefore, the sources of PAHs of the houses came from incense burning along with the traffic. The elevated PAH levels might come from human activities, moreover, the traffic volume around that area was investigated to be the main source of PAHs for industrial and its vicinity.

Figure 6 Diagnostic ratios of indoor PAHs (a) industrial area and (b) non-industrial area.

Figure 7 Diagnostic ratios of ambient PAHs.

For ambient air analysis, the result of diagnostic ratios showed that the major source of outdoor PAHs was the transportation sector followed by petrol emission (Figure 7). The types of PAHs depended on several factors including the types of fuel and stability of substances. Normally, particle-bond PAHs were predominantly produced from solid and liquid fuel combustion; hence, the diagnostic ratios expressed that the traffic was a major source of indoor PAHs. Likewise, the industrial fuel was generally used in a gas phase so the PAHs ratio

from this source was lower than that released from the traffic source. The diagnostic ratios of PAHs from other studies are expressed in Table 3.

Conclusions

Several house characteristics were observed for both industrial and vicinity areas in this study. Each house has different residential activities (burning incenses and cooking using biomass fuels) that can cause both PM_{10} and PAHs emission. Daily average (24 h) ambient

air concentrations of PM_{10} mainly relied on the traffic volume. In contrast to ambient air, the indoor PM10 levels at six different communities were slightly varied with the residential activities and the amount of vehicle in that area. On account of PAH concentrations, BBF was the major species among all PAH compounds in all communities for both indoor and ambient air. Mostly, PAHs derived from the incomplete combustion and traffic density and these factors were the pollutants of the greatest concern. Furthermore, the levels of PAHs in the industrial communities were greater than those calculated for the vicinity communities. The household activities (cooking and incense burning) along with traffic volume of each sampling site were played an important role in this study. For that reason, the management of these sources of both PM_{10} and PM_{10} -bound PAHs should be strengthened, also further monitors should be required in the industrial area and its vicinity. For source identification of PAHs, the limitations of the diagnostic ratios might result in the unreliability of data interpretation, mainly for LWM PAHs. Hence, it is suggested that the consideration of this process should be investigated in future studies.

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