



DISTRIBUTION OF POLYCYCLIC AROMATIC HYDROCARBONS IN SURFACE SEDIMENTS OF KOH SICHANG ANCHORAGE AREA IN THAILAND

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The levels and distribution of polycyclic aromatic hydrocarbons (PAHs) were investigated in surface sediments of Koh Sichang anchorage area of Thailand using gas chromatography with flame-ionization detection (GC/FID). The total concentrations of 16 PAHs US EPA priority pollutants varied from 65.2 to 18,970 ng g⁻¹ dw, with a median concentration of 282±3,660 ng g⁻¹ dw. The sediment samples were classified as moderately contaminated compared to those observed in other regions. PAH compositional signatures of surface sediments of the study area were dominated by higher molecular weight PAHs (4-6 rings) comprising about 87 per cent of total PAHs concentrations. Dibenzo[a,h]anthracene, benzo[b]fluoranthene, benzo[g,h,i]perylene, benzo[a]pyrene, fluoranthene, indeno[1,2,3-cd]pyrene, and pyrene represented the highest fractions in most surface sediment samples. Source apportionment using diagnostic PAH ratios indicated that composition of PAHs in most sediment samples originated mainly from incomplete combustion of organic matter (pyrolytic origin), with a mixture of pyrolytic and petrogenic PAHs were observed in some of the study sites. The presence of almost all human carcinogenic PAHs in the study area indicated that these sediments can be considered contaminated sites, suggesting that future monitoring programs together with an effective coastal management program must be implemented to ensure health and safety for all.

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sediment or soil particles. Owing to their aromatic structure, PAHs are inherently stable in the environment, particularly under reducing conditions, such as encountered in sediments.⁵ Even under favourable conditions, the sorbed PAHs will be released to the water as an extended source to threaten the aquatic ecosystem through bioaccumulation in food chains.⁶ Thus, understanding the contributions of the various sources is essential and important for appropriately managing PAH levels in the environment. However, our knowledge of PAH contamination in Thailand has been limited.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of diverse organic compounds containing two or more fused aromatic rings of carbon and hydrogen atoms. They are widespread contaminants in marine, river and lake sediments and are always found as a mixture of individual compounds. PAHs enter the environment by a variety of pathways. They are present in unburned petroleum (petrogenic PAHs) and can be released directly to the environment both by human activities (oil spill) and natural processes (oil seepage). Diagenetic processes are also suspected to generate certain PAHs (e.g. perylene) from biogenic precursors. In general, biosynthesis is considered a localized source, with little impact on global concentrations. The most prominent and ubiquitous source of PAHs to the environment is the incomplete combustion of modern (wood) and fossil (petroleum and coal) biomass.^{1,2} High-molecular-weight PAHs from anthropogenic sources can reach toxic concentrations that are detrimental to the environment and human health. Several PAHs are known to be carcinogens and/or mutagens or precursors to carcinogenic sister compounds.^{3,4} Accordingly, they had been included in the USEPA and the EU priority monitoring pollutants list. PAH compounds persist in the environment and, due to their hydrophobic nature, become associated with particulate matter, and preferentially concentrate on

The presence of PAHs in the marine environment has attracted the attention of the scientific community as these compounds are frequently detected in sediments at increasing levels and many of these compounds are potential or proven carcinogens, hence finding the sources of PAH contamination are commonly of environmental concerns. The major sources of PAHs may be either natural or anthropogenic. There are two anthropogenic PAH sources: pyrolytic hydrocarbons from combustion sources and petrogenic hydrocarbons from petroleum sources. Pyrolytic PAHs, characterized by a predominance of parent compounds with four or more aromatic rings, can be emitted from the incomplete combustion of fossil fuel, urban and industrial activities, natural fire and biomass burning that produce high molecular weight and less or non-alkylated PAHs.^{1,2,7} Combustion sources of PAHs in the industrialized countries include power plants, refineries, and automobiles.⁸ Petrogenic PAHs, compounds with two to three aromatic rings, are mainly derived from the release of crude oil and petroleum products such as lubricating oil, diesel fuel, gasoline, asphalt and kerosene.^{9,10} In this respect, the molecular indices based on the ratio of selected PAH concentrations in sediments can be used to elucidate the possible sources. The ratios between low and high molecular weight PAHs⁹ and those of specific compounds, such as Flt/(Flt+Pyr), BaA/(BaA+Chr), Flt/Pyr and BaA/Chr have been proposed as valuable source indicators.¹⁰ In

addition, diagnostic ratios of BaA/Chr, BbF/BkF, BaP/BeP and InP/BgP have been applied to identify specific types of combustion, such as vehicle exhaust, coal/coke combustion, forest fires, and smelters.¹¹⁻¹³

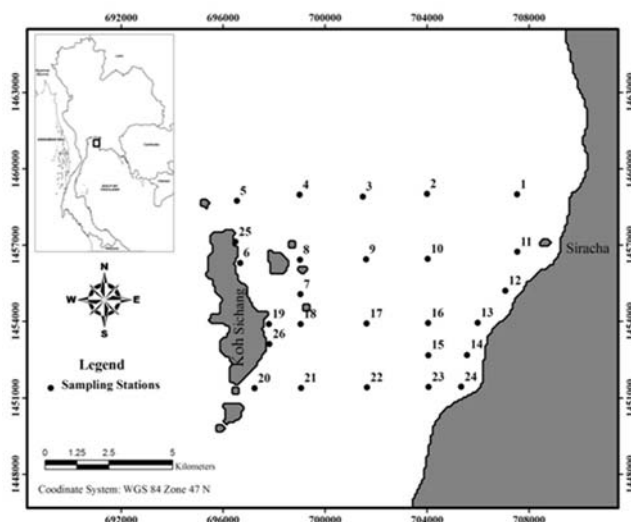


Figure 1. Map of Koh Sichang and Siracha anchorage area in the inner Gulf of Thailand showing the sample collection sites.

Koh Sichang (Sichang Island) is a small island located along the eastern coast of the inner Gulf of Thailand (latitude 13°7.4' - 13°11.5'N, longitude 100°49.1' - 100°54.9'E), twelve kilometres offshore from the town of Siracha, Chonburi Province (Fig. 1). Due to its proximity to shipping lane, the marine area between Koh Sichang and Siracha has made a convenient anchorage spot for dozens of barges which tranship their cargoes to lighter boats for the trip up the Chaophraya River to the Bangkok Port. The transshipment activities of coal in the area has resulted in an increase of fine coal dust and particles which will eventually settle down and act as a likely source of PAHs into the ambient environment. The area also experienced more than twenty oil spill events during the last two decades.¹⁴ In addition, human activity along Siracha and Koh Sichang coasts has resulted in a number of possible point sources of contamination; including a variety of industry activities, an oil refinery, ports, ferry terminals and fishing piers, being discharged into the receiving environment. Accordingly, this ecosystem during the years has accumulated in sediments both elevated levels of organic matter and petroleum hydrocarbons;¹⁴ Hence it is necessary to investigate the pollution sources and their environmental impacts on this coastal marine environment.

The objective of the present work was to quantify and determine the distribution of the 16 US EPA priority pollutant PAHs in sediments from the Koh Sichang anchorage area. In addition, the possible sources of PAH contamination were studied using PAH diagnostic ratios. The study will provide valuable information to be referenced by engineers, planners and officials for future management strategies on PAH contamination in this area.

Experimental

Sample collection

Surface sediment samples (0–3 cm depth) were collected by using modified Van Veen grab sampler in February 2012; from 26 sites distributed along four transect lines in the coastal area of about 100 square kilometres between Koh Sichang and Siracha (Fig. 1). Two replicate grabs from each site were mixed thoroughly in a bowl to provide a single composite sample for each site. Immediately after collection, the samples were scooped into aluminium boxes, which have been pre-washed with n-hexane and kept in an icebox, and then transported to the laboratory, where they were divided for physico-chemical analysis and for further PAH assessment. The considered physico-chemical parameters included the analysis of percentage water content, percentage organic carbon, and sediment characterization while sediments for PAH analysis were kept frozen until further processing. Prior to extraction sediments were thawed at room temperature, mixed thoroughly, freeze dried and sieved through a 250 µm sieve to remove coarse substances.

Analytical methods

Samples were analysed for PAHs using the procedure of IAEA/IOC/UNEP,¹⁵ with slight modification. About 50 g of the dried sediment sample was carefully transferred into the extraction thimble and placed in the extraction chamber of the Soxhlet extraction unit. An internal standard (9,10-dihydroanthracene) was added, and the sample was Soxhlet extracted with methylenechloride for 12 h. The extract was reduced in volume, cleaned up and fractionated on a silica gel-alumina column,¹⁶ reduced in volume again with a gentle stream of ultra-pure nitrogen until 0.5 mL, and finally the extract was analysed by using capillary gas chromatography.

PAHs were analysed with Agilent 6890 gas chromatography coupled with flame ionization detector (FID) system equipped with a fused silica capillary column coated with HP-5 5 % phenylmethylsiloxane (30 m length and 0.32 mm ID 0.25µm film thickness). A 1.0 µL aliquot of the extract was injected while the injector port was held at 250 °C and operated in splitless mode. The oven temperature program started at 70°C covered a range from 70 °C to 290 °C at 6 °C min⁻¹ with 15 min hold. Helium was used as carrier gas with flow rate at 1.0 mL min⁻¹ constant flow and detector temperature was 300 °C. Hydrogen and air ratios were optimized and their values were chosen as 30 mL min⁻¹ and 300 mL min⁻¹ respectively.

The following 16 PAHs were quantified: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Acn), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (InP), dibenzo[a,h]anthracene (DBA), and benzo[g,h,i]perylene (BgP). Total PAHs (Σ PAHs) was the sum of the 16 PAHs.

Table 1. Mean(\pm sd) and range of the measured physico-chemical parameters of sediments of Koh Sichang anchorage area.

	% Sand	% Silt	% Clay	Texture	% Water	%OC	pH	E_h (mV)
Mean \pm s.d.	26.9 \pm 19.7	47.7 \pm 17.1	25.4 \pm 5.7	silty clay	48.4 \pm 13.2	1.5 \pm 0.6	7.5 \pm 0.2	-79 \pm 55
Range	4.3-80.8	1.9-65.5	15.5-39.2		21.7-65.3	0.4-2.5	7.2-8.0	55-(-187)

Compound concentrations below detection limits were assumed to be zero for the summation of Σ PAHs in each sample. The identification of PAHs was based on comparison of the retention times of the peaks with those obtained from standard mixture of PAHs (Supelco Ltd.) and from spiked samples analysed under the same conditions. Quantification was based on external calibrations curves prepared from the standard solution of each of the PAHs. The coefficients of determination (r^2) for the PAH standard calibration plots were Nap (0.994), Acy (0.997), Acn (0.993), Flu (0.994), Phe (0.996), Ant (0.996), Flt (0.992), Pyr (0.998), BaA (0.992), Chr (0.995), BbF (0.991), BkF (0.990), BaP (0.990), InP (0.991), DBA (0.994), and BgP (0.991). All the analysis was carried out in three times. The relative standard deviations ranged from 0.07 to 0.45 %.

Analysis of procedural blanks and spiked samples with each set of analysed samples was used to assess quality control measurements. Four deuterated PAH surrogates (d_8 -naphthalene, d_{10} -fluorene, d_{10} -fluoranthene, d_{12} -perylene) were added to the samples and matrix blank prior to extraction. Recoveries of surrogates generally ranged from 80 % to 105 % of the spike concentration, with the overall mean of recovery 85 ± 8.2 %. Concentrations of PAH compounds were not corrected for surrogate recoveries, and are expressed on a dry-weight (dw) basis.

Results and Discussions

Sediments characteristics

The results of sediment characteristics are presented in Table 1. The grain size was utilized to analyse the types of sediments. Sieve analysis and fine grain analysis show the majority of the sediments consisted of a silt mixture. The average mean grain size of surface sediments for most of the studied sites was silty clay. The organic carbon (OC) content ranged from 0.4 % to 2.5 %, with the average value of 1.5 ± 0.6 %. Sites 7, 8, 25 and 26 had the highest OC contents, which is expected since these sites are either close to the coal transshipment area (7 and 8) or located near the ferry terminals (25 and 26). Sites 1 and 6, had the lowest OC contents due to more sand-sized material (>65 %) in the sediments.

Distribution and composition of PAHs

The Σ PAHs concentrations in surface sediments of the Koh Sichang-Siracha anchorage area ranged from 65.2 ng g^{-1} to $18,970 \text{ ng g}^{-1}$, with a median concentration of

$282\pm 3,660 \text{ ng g}^{-1} \text{ dw}$. The highest Σ PAHs concentration was found at site 26, which is the nearest site to a ferry terminal and ship repairing facilities. The second highest concentration was found at site 7 (1.795 ng g^{-1}), which is the coal transshipment area. The Σ PAHs concentration detected in sediment sample of site 26 is around 290 times higher than the lowest level detected at site 16. Based on the PAHs levels suggested by Baumard et al.,¹⁷ most sediment samples in the study area can be classified as moderately contaminated (Σ PAHs = $100\text{-}1,000 \text{ ng g}^{-1} \text{ dw}$). The Pearson correlation coefficients for Σ PAHs concentration and sediment characteristics in the study area were carried out. Results show that the sediment Σ PAHs concentrations were not significantly correlated to either OC or particle size ($p > 0.05$). The observation suggests that OC and particle size are not major factors to control the Σ PAHs distribution.

Relative abundance of each PAH compound

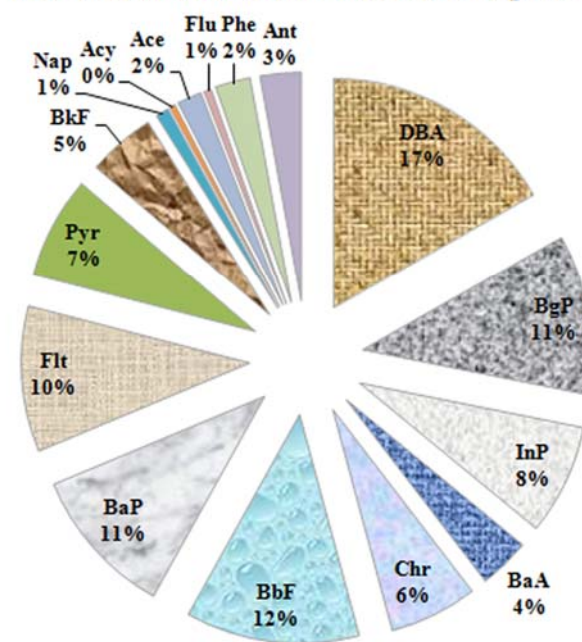


Figure 2. The percentage of mean concentration of each PAH compound in the study area.

The percentage of mean concentration of each PAH compound in the study area is shown in Fig. 2. Many of the PAH compounds were present at low concentration range. The individual PAH median concentrations ($\text{ng g}^{-1} \text{ dw}$) found in the study area were in the order: DBA (45.5) > BbF (33.3) > BgP (30.9) > BaP (28.8) > Flt (28.0) > InP (21.2) > Pyr (19.6) > Chr (16.9) > BkF (13.0) > BaA (9.9) > Ant (7.8) > Phe (6.7) > Acn (4.7) > Nap (2.5) > Flu (1.4) > Acy (1.1). The difference in PAHs abundance in surface

sediments may be attributed to molecular weight and microbial degradation. A wide varieties of microorganisms including bacterial, fungal, and algal strains are known to degrade PAHs. Lower molecular weight PAHs such as Nap and Phe are degraded rapidly in sediments, but higher molecular weight PAHs such as Pyr, Flt, BaA and BaP are more resistant.¹⁸ Hence, the distribution of PAHs found in sediments gives information about the precursor sources.

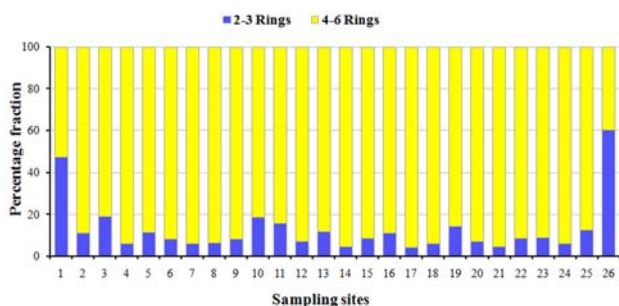


Figure 3. Relative distribution of % (2-3)-rings and % (4-6)-rings PAHs in Koh Sichang - Siracha surface sediments. 2-3-rings: Nap, Acy, Acn, Flu, Phe, Ant; 4-6-rings: Flt, Pyr, BaA, Chr, BbF, BkF, BaP, InP, DBA, BgP.

PAHs composition in the sediments of the Koh Sichang-Siracha anchorage area were generally similar in such a way that the relative contribution of low molecular weight (LMW) components with 2-3 rings (LPAHs) was lower (ranging from 4 % to 19%), while components with high molecular weight (HMW) with 4-6 rings (HPAHs) were dominant, ranging from 81 % to 96 %, except the sediments from sites 1 and 26, where the 2- & 3-ring PAHs were predominant at 47-60 %, respectively (Fig. 3). The result suggests that the PAHs contamination in these two sites comes from a different source than those for the PAHs found in other locations. The spatial distribution patterns of Σ PAHs, Σ LPAHs and Σ HPAHs in the study area shown in Fig. 4 reveal the predominance of high molecular weight PAHs in the sediments which reflects the presence of significant combustion products from pyrolytic processes and/or pyrolytic sources.^{1,2,7}

PAH diagnostic ratios

Several PAH isomeric ratios have been used to identify different sources of PAHs to environmental samples. These ratios are useful indicators of PAH sources, and have been widely used to infer the source of PAHs found in sediments.^{10-13,19} For example, ratios of Phe/Ant and Flt/Pyr have been widely used to distinguish petrogenic and pyrogenic (pyrolytic) sources of PAHs.^{10-13,19-22} It is generally accepted that PAHs with Flt/Pyr values >1 are related to pyrolytic origins and values <1 are attributed to petrogenic origin. PAHs of petrogenic origin are also characterized by Phe/Ant >10 , while combustion process often result in Phe/Ant <10 . Other common ratios that have been used include Ant/(Ant+Phe) and Flt/(Flt+Pyr).²³⁻²⁸ Ratios of Ant/(Ant+Phe) <0.1 and Flt/(Flt+Pyr) <0.4 usually imply a petrogenic source, whereas ratios of Ant/(Ant+Phe) >0.1 and Flt/(Flt+Pyr) >0.5 suggest a pyrogenic source and combustion source of biomass (grass, wood, or coal combustion), respectively.

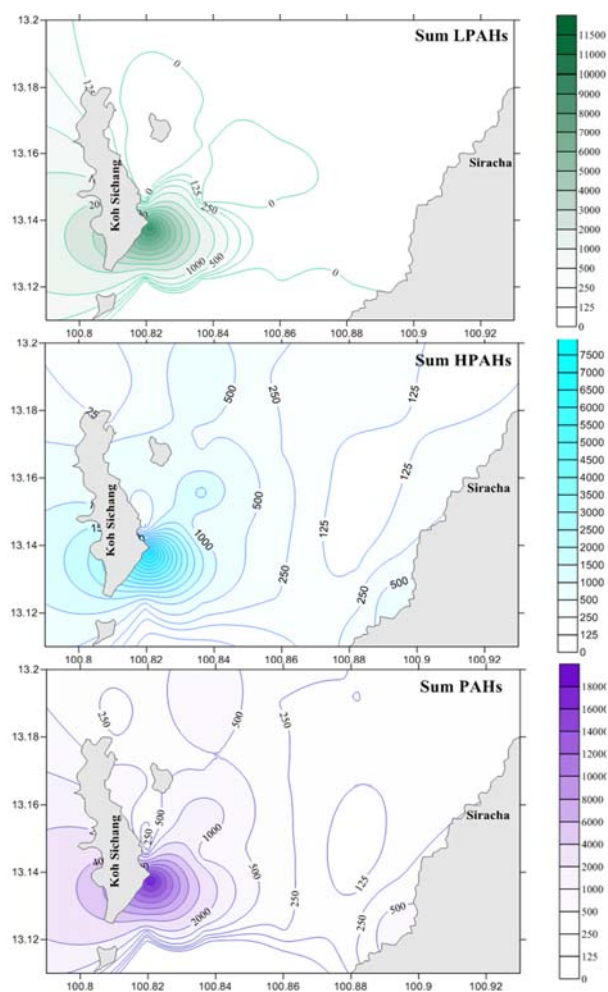


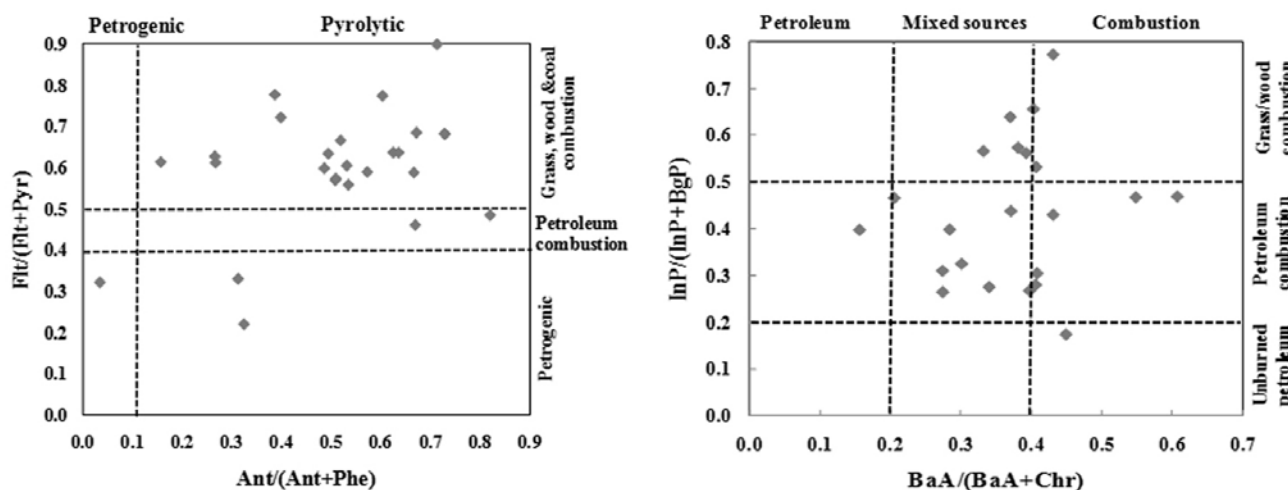
Figure 4. The spatial distribution of Σ LPAHs, Σ HPAHs and Σ PAHs concentrations ($\text{ng g}^{-1} \text{dw}$) in the study area.

Flt/(Flt+Pyr), BaA/Chr, BaA/(BaA+Chr), InP/(InP+BgP), Phe/Ant and Flt/Pyr were calculated for the sediment samples in the study area in order to determine probable PAH sources (Table 2). Ratio of different isomer pairs were plotted for use to identify PAH sources as shown in Fig. 5. At most of the sites investigated in this study, the Ant/(Ant+Phe) ratios were generally >0.1 , ratios of Flt/(Flt+Pyr) were >0.4 , ratios of InP/(InP+BgP) >0.2 , ratios of Phe/Ant >10 , ratios of Flt/Pyr >1.0 . These indicate that pyrolytic sources were the primary sources of sedimentary PAHs in the study area. However, the BaA/Chr and BaA/(BaA+Chr) ratios indicated the mixed petrogenic and pyrolytic sources of the sedimentary PAHs.

Fig. 5 shows the cross plots for the ratios of Flt/(Flt+Pyr) vs. Ant/(Ant+Phe) and InP/(InP+BgP) vs. BaA/(BaA+Chr) in sediments, which indicated that most sediment samples have mixed combustion sources. Several of the Koh Sichang-Siracha sediment samples, however, have InP/(InP+BgP) ratios indicated that the main sources of PAHs were combustion of petroleum, while several sediment samples have InP/(InP+BgP) ratios indicated that of grass or wood combustion source.

Table 2. The diagnostic PAH source ratio guidelines* and those obtained for surface sediments in the study area.

Diagnostic ratios	Petrogenic source	Pyrolytic source	This study	
			Mean \pm sd	Range
Ant/(Ant+Phe)	<0.1	>0.1	0.51 \pm 0.19	0.03 – 0.82
Phe/Ant	>15	<10	2.27 \pm 5.47	0.22 – 28.52
Flt/Pyr	<1.0	>1.0	1.91 \pm 1.61	0.28 – 8.86
Flt/(Flt+Pyr)	<0.4	>0.4	0.6 \pm 0.15	0.22 – 0.90
BaA/Chr	<0.4	>0.9	0.60 \pm 0.31	0.04 – 1.55
BaA/(BaA+Chr)	<0.2(0.2-0.35)	>0.35	0.35 \pm 0.12	0.04 – 0.61
InP/(InP+BgP)	<0.2	>0.2	0.43 \pm 0.15	0.17 – 0.77

*Modified from Yunker *et al.*^{12, 28}**Figure 5.** PAH cross plots for the ratios of Flt/(Flt+Pyr) vs. Ant/(Ant+Phe) and InP/(InP+BgP) vs. BaA/(BaA+Chr) in sediments of Koh Sichang-Siracha marine area.

Conclusion

This study provides important data set on PAH levels in surface sediments of Koh Sichang-Siracha marine area in Thailand. The PAH concentration levels of 16 PAH priority pollutants ranged from 65.2 to 18,970 ng g⁻¹ dw with a median concentration was 282 \pm 3,660 ng g⁻¹ dw. The overall levels of PAHs were moderate compared to coastal areas in other regions. The PAH distribution profile indicated potential source dependence, as the levels were generally higher in the vicinity of known inputs such as coal transshipment area, ferry terminals and ports. PAH diagnostic ratios indicated that PAHs in surface sediments mainly from pyrolytic sources (i.e. derived from combustion of petroleum and other organic materials) and very similar to PAH signatures of many coastal marine sediments elsewhere. The pyrolytic part of the pattern could arise from atmospheric transported coal derived particles from coal transshipment area, ship/boat emissions and other combustion processes. At most of the sites investigated in this study, the sedimentary PAH concentrations were dominated by DBA, BbF, BgP, BaP, and InP, which are IARC probable and possible human carcinogens.³ Hence, high contaminated sediment samples in the study area are expected to have a high toxic potential. At present, no criteria or standards have been set for PAHs by any regulatory agency for the protection of sensitive species of aquatic organisms in Thailand. It is recommended that

detailed assessment of ecological and human health risks associated with these compounds should be carried out as a matter of priority.

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