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Petroleum pollution in the Gulf of Thailand: A historical review

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Abstract—With its rapid economic growth in the past three decades, Thailand is encountering serious environmental problems. Among these, the issue of environmental pollution caused by oil has become increasingly important because their usage in large quantities have resulted in negative impacts on marine and coastal ecosystems including health effects on wildlife. Major oil spills are of public concern and usually under control of pollution authorities. However, small oil spills occurring in offshore areas due to ship operations are usually uncontrolled, unaccounted in oil spill balance and have smaller public concern. A review is presented of the database generated to the present concerning petroleum hydrocarbons in waters and sediments of the Gulf of Thailand. Historical changes in the extent of contamination are discussed and assessed by comparison to data produced elsewhere. In general, the level of petroleum contamination in the Thai waters is low to medium level compared with more urbanized and industrialized countries. Nevertheless, continuous monitoring and investigations on the level of petroleum contamination are still needed in Thailand.

Key words: petroleum hydrocarbons, tar ball, DDPH, PAHs, Gulf of Thailand

Introduction

The Gulf of Thailand is a semi-enclosed tropical sea located in the South China Sea (between latitudes 5°00′N and 13°30′N; longitudes 99°00′E and 106°00′E), surrounded by the countries Malaysia, Thailand, Cambodia and Vietnam. The Gulf covers roughly 320,000 km^2. It is relatively shallow; the mean depth is 45 m, and the maximum depth only 80 m. The general shape of the Gulf's bottom topography can be considered elliptic parabolic. It is separated from the South China Sea by two ridges that limit water exchanges with the open South China Sea. The first extends southeast from Cape Camau for about 60 nautical miles with an average sill depth of less than 25 m. The second ridge, which extends off Kota Bharu for approximately 90 nautical miles, has an average sill depth of 50 m. There is a narrow, deeper channel between the two ridges with the observed depth of 67 m (Emery and Niino 1963). The Gulf may be divided into two portions, Upper Gulf and Lower Gulf. The Upper Gulf, at the innermost area has an inverted U-shape, is located between latitudes 13°30′N and 12°60′N and longitudes 100°00′E and 101°00′E (Fig. 1). It is the catchment basin of four large rivers on the northern side and two on the western coast. Among them, the Chaophraya River has the biggest volume transport next to the Mekong River. The average runoff per year of the Chaophraya is 13.22×10^3 MCM and that of the Mekong is 326×10^3 MCM (Piyakarnchana et al. 1990). Seasonal circulation in the Gulf of Thailand, deduced from oceanographic data measured in 1993-1994 (Wattayakorn et al. 1998) and from monthly-mean winds obtained from meteorological stations around the Upper Gulf during 1980–2000 (Buranaprathepprat et al. 2006) suggested that circulation in the Gulf is generally weak and variable. In the Upper Gulf, the counter-clockwise circulation is apparent during the northeast monsoon. However, both clockwise and counter-clockwise circulations may occur during the southwest monsoon depending on wind condition and external flow through the open boundary. On the whole, the Gulf of Thailand is poorly flushed. In the Upper Gulf, little mixing occurs between coastal and offshore waters. As a consequence of these comparatively static conditions, land-based contaminants discharged into the Upper Gulf may accumulate while variability in current directions may also result in the return of contaminants that were initially flushed from an area (Wattayakorn et al. 1998).

Thailand's population increased from 23 million in 1961 to about 67 million by 2009. The rapid increasing population with associated industrialization and economic development in the coastal areas in the past two decades has resulted in construction and planning of many coastal development projects. The Port of Bangkok, at the mouth of the Chaophraya River, is the transportation life-blood of the country. It serves as the main egress point for agricultural export products from the country's interior as well as access for raw materials and oil products. In addition, several deep sea ports within the Gulf were constructed to facilitate the huge increase in sea transport of import and export cargoes. The deep sea port at

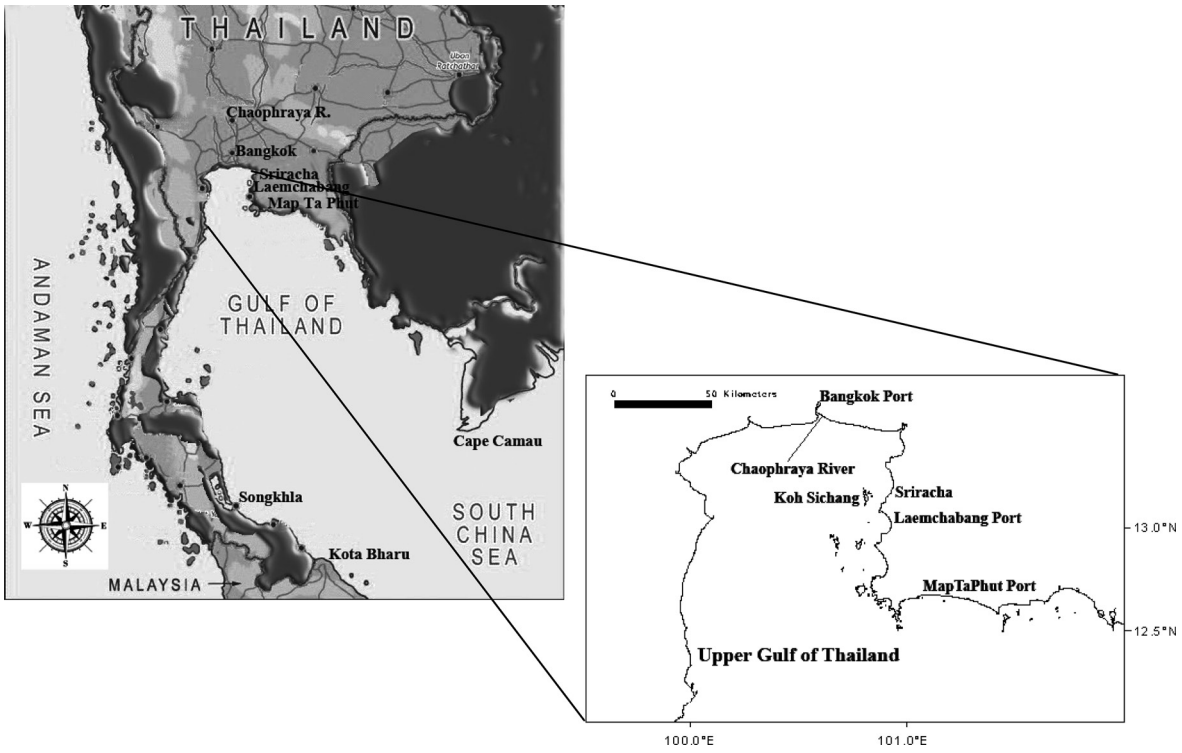

Fig. 1. Map showing the Gulf of Thailand, major seaports and the Chaophraya River.

Laemchabang provides relief to the congestion in Bangkok, while the ports at Songkhla and Map Ta Phut offer ocean shipping alternatives to industries and resources within the Gulf of Thailand. Many other smaller coastal ports have been constructed for fishing fleets and coastal freighters to serve as feeder ports. There are seven oil refineries in Thailand: Chevron (Star), Esso, Bang Chak, Thai Oil, IRPC, PTT Aromatics and Rayong Pure, with a total daily refining capacity of 1.2 million barrels. The three main refineries are located on the eastern seaboard of the Gulf, i.e. Shell Co. of Thailand Ltd. (275,000 bbl/d) located in Rayong, Thai Oil Co. Ltd., in Sriracha (192,850 bbl/d) and Esso Standard Thailand Ltd. (173,500 bbl/d), also located in Sriracha. The accidental oil spills have been frequently reported along oil transport routes, at points of discharge and loading of oil carriers. There were over 200 oil spill incidents reported to occur in the Thai waters during 1974–2009. Frequent spills were found at the mouth of the main entrance to the Bangkok Port (Chaophraya River mouth), Koh Sichang-Sriracha (Chonburi), Laemchabang Port (Chonburi) and Map Ta Phut Port (Rayong). These oil spills represent the greatest source of petroleum related pollution in the Gulf and are of public concern. However, many small oil spills do occur in the open sea due to ship operations which are usually uncontrolled and have smaller public concern. Figure 2 shows the number of oil spills incidents recorded in the Thai waters from 1974 to 2009 (Marine Department 2009), while Table 1 lists the major spills events in the Thai waters. There is an increasing trend of oil spills over the last two decades. Aside from oil

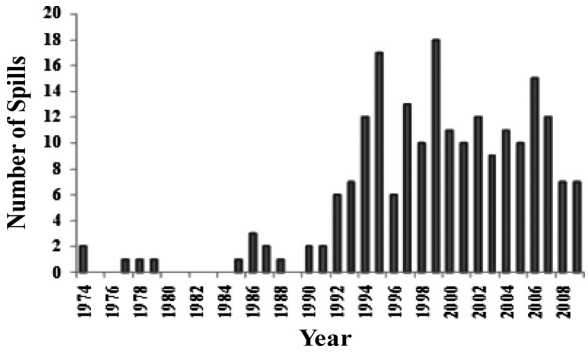

Fig. 2. Frequency of major accidental oil spills in the Thai waters.

spills through accidents, discharges of oil also occur from operational shipping activities; such as deballasting, tank cleaning, dry docking, bunkering, cargo loading and unloading. Deballasting in the South China Sea is more probable and frequent due to frequency of tankers traversing the water. Oil released during deballasting operations is in the form of tar lumps and weathering produces tarry residue and tarballs. Since the Gulf of Thailand receives water from the current circulation in the South China Sea during the yearly monsoon (Wyrtki l961, Wattayakorn et al. 1989), this might possibly explain the stranded tarballs found on the beaches around the Gulf (Piyakarnchana et al. 1979, Intharapanich 1979, Petpiroon et al. 1986, Suthanaruk 1991, Intang 2006). In general, tarballs accumulations and locations vary seasonally and in close relation to the current circulation and tanker

Table 1. Major oil spill incidents in the Gulf of Thailand from 1973 to 2009.

Date	Oil type	Volume (tonnes)	Location	Cause
10 Apr 1974	Diesel & Fuel oil	2100	Chaophraya River mouth	Collision of tanker (Visahakit) and container vessel
29 May 1977	unknown	300	Chaophraya River mouth	Collision of tanker (Vachira) and container vessel
1979	Fuel oil	300	Koh Sichang, Chonburi	Grounding due to fire (Sun Flower)
6 Mar 1994	Diesel oil	400	Koh Sichang, Chonburi	Collision of tanker (Visahakit5) and container vessel
16 Jan 1996	Fuel oil	200	Chaophraya River mouth	Collision of container vessels
30 Oct 1996	Crude Oil	150	Map Ta Phut, Rayong	Leaking during loading
15 Jan 2002	Diesel oil	240	Sattahip, Chonburi	Grounding of tanker (Eastern Fortitude)
17 Dec2002	Fuel oil	210	Laemchabang, Chonburi	Collision of tanker (Sky Ace) and container vessel
26 Dec 2004	Fuel oil	150	Koh Larn, Pattaya, Chonburi	Grounding of tanker (Dragon1)

routes (Kurashina 1975, Nasu et al. 1975, Limpsaichol 1984).

This review paper summarizes available information on the studies of petroleum hydrocarbon contamination in waters and sediments of the Gulf of Thailand.

Petroleum hydrocarbon contamination

Petroleum pollution of the marine and coastal environment in Thailand is widely recognized as a problem and has been noted at various national and regional meetings over the last decades. Numerous studies and monitoring activities concerning petroleum hydrocarbon contamination in marine and coastal waters have been carried out in Thailand by various academic institutions since the mid-1970s. Most studies and monitoring activities were conducted in estuarine and nearshore waters, with relatively fewer studies conducted in offshore water. Early studies on petroleum hydrocarbon contamination were carried out during the Marine Pollution (Petroleum) Monitoring Pilot Project (MAPMOPP) of the Integrated Global Ocean Survey System (IGOSS) which was implemented in Thailand in 1976 until 1980. The relatively simple technique of UV fluorescence spectroscopy (UVF) was employed to quantify the dissolved/dispersed petroleum hydrocarbons (DDPH) and petroleum hydrocarbon content in sediments, using chrysene as the standard reference compound according to the MAPMOPP protocol (IOC/IMO 1976). Emission scans and synchronous excitation/emission scans were recorded to evaluate the relative fluorescence and to characterize the polycyclic aromatic hydrocarbons (PAHs). This fluorescence includes not only contributions from PAHs but also from metabolite PAHs. Synchronous spectra indicate the presence of a range of PAHs including mono-, bi-, tri- and tetracyclic aromatics. Unlike many aliphatic hydrocarbons, PAHs are a component of crude oils that have not been found to be produced biogenically, and their presence in marine samples is therefore taken to indicate the presence of petroleum hydrocarbons (Farrington and Meyer 1975, Law 1981). Owing to its ease of operation, rapidity, and sensitivity, the UVF technique has been adopted for many national and international monitoring programs (Abu-Hilal and Khordagui 1994, Abdullah et al., 1994, 1996, Chumchuchan et al. 1998, Marrugo González et al. 1999, Zanardi et al. 1999, Shriadah 2000, Hii et al. 2002, Law et al. 2004, Chouksey et al. 2004). This technique has also been widely used for monitoring petroleum hydrocarbon concentrations in seawater (according to Ehrhardt 1983, IOC/UNESCO, 1984 and Parsons et al. 1984), and sediment samples (IOC/UNEP/IAEA 1992) in Thailand. Concentration ranges of petroleum hydrocarbons found in the water and sediments during each study period are shown in Table 2.

A typical example of hydrocarbon distribution in seawater along the eastern coast of the Upper Gulf of Thailand is shown in Fig. 3. Hydrocarbons present in the marine environment are actually originated from various sources. Ship-breaking industry was considered a major source of petroleum to seawater along the Eastern Seaboard of the Gulf of Thailand. Suthanaruk (1991) reported that during the scraping period petroleum hydrocarbon concentrations in the offshore sea surface microlayer (about 7 km from the ship-breaking activities) ranged from 4.1 to 31.8 $\mu g\,l^{-1}$ were found. Enrichment factors were reported to range from 6 to 145 times relative to concentrations occurring in the underlying subsurface seawater. Owing to the substantial amount of oily water discharged from land-based sources and boating activities near shore, the poor water flushing in the bay area and characteristics of current movement, high hydrocarbon levels are always found in the inshore waters associated with port/pier areas with a higher concentration of maritime activities and the transportation and offloading operations of crude oil at the terminals of refineries. Coastal waters along recreational beaches, on the other hand, were found to have significantly lower concentrations of hydrocarbons (Chumchuchan et al. 1998). Generally, higher levels of petroleum hydrocarbons were reported in estuarine and coastal waters of the inner Gulf than that in the offshore water of the Lower Gulf.

A study conducted by Wattayakorn et al. (1998) on the seasonal dispersion of petroleum hydrocarbon levels in the

Table 2. Ranges and mean concentrations of petroleum hydrocarbons in water and sediments at various study areas in the Thai waters, as determined by UVF spectroscopy technique.

Study areas	Water ($\mu g\, l^{-1}$) chrysene equivalent	Sediments ($\mu g\, l^{-1}$ dw) chrysene equivalent	References
		Marine and coastal areas	
Offshore waters:	0.37–1.42	0.01–0.50	Intharapanich, 1979
Gulf of Thailand	0.05–4.13 (0.68±1.08)	n.d.	Wongnapapan et al., 1999
Upper Gulf	0.06–6.10	0.06–4.20	Sompongchaiyakul et al., 1986
Upper Gulf	0.17–0.89	n.d.	Chartkittikunwong, 1986; Onodera et al.,1987
Upper Gulf	0.65–8.3*	0.70–62.0*	Wattayakorn, 1986;
Lower Gulf	0.07–6.6*	0.03–8.30*	Wattayakorn, 1987a & b
	*crude oil equivalent		
Coastal waters:			
Upper Gulf	0.20–8.26		
Lower Gulf	0.01–12.0	n.d.	Wattayakorn et al., 1997,1998;
Eastern coast	0.05–11.8		Chumchuchan et al., 1998
Andaman Sea	0.06–2.64		
Eastern coast:	6.19–14.57	0.38–1.65 (wet weight)	Petpiroon et al., 1986
(Chonburi to Trad)	0.01–5.29	n.d.	Petpiroon, 1988
Rayong & Trad	0.14–6.72	n.d.	Kan-atireklap et al., 2005
Rayong	0.05–11.8 (1.38±0.86)	n.d.	Tappatat, 1995
Ao Phe, Rayong	0.61±0.26	1.53±2.39	Sanguansin et al., 2003
Chonburi & Rayong	0.04–48.1 (during oil spills)	n.d.	Kan-atireklap et al., 2007
Sattahip, Chonburi	0.03–0.29 (0.11±0.08)	0.39–13.0 (5.69±3.55)	Sri-on, 2006; Intang, 2006
Sriracha, Chonburi	0.10–12.5 (0.89±1.48)	1.10–153.4 (29.4±30.0)	Wattayakorn & Rungsupa, 2011
Koh Chang & Koh Kood, Trad	0.85–6.61	n.d.	Kan-atireklap et al., 2002
Koh Sichang, Chonburi	0.60–5.43 (2.42±1.61)	n.d.	Rungsupa & Wattayakorn, 2008
Koh Sichang, Chonburi	0.10–12.5 (0.89±1.48)	1.10–153.4 (29.4±30.0)	Wattayakorn & Rungsupa, 2011
Fishing pier:			
Bangsan & Angsila, Chonburi	0.96–11.2 (4.81±3.37)	0.37–23.5 (6.82±8.22)	Meepoka, 2007
Angsila & Sriracha, Chonburi	5.76–13.8 (9.9±1.9)	n.d.	Chomchuen, 2008
Ship-breaking area:	DDPH: 0.02–19.4 (1.05±1.33)		
Map Ta Phut, Rayong	Surface micro-layer: 0.52–31.8 (7.05±5.68)	0.06–1.86 (0.50±0.36)	Suthanaruk, 1991
		Rivers/Estuaries	
Chaophraya estuary	2.87–43.8 (12.1±10.6)	n.d.	Nokyoo, 1995
Thachin estuary	0.93–4.25 (2.07±0.88)	4.83–151.2 (52.8±37.4)	Sunwanich, 1991; Sunwanich & Wattayakorn, 1991; Wattayakorn & Sunwanich, 1992

Table 2. (Continued).

Study areas	Water ($\mu g\,l^{-1}$) chrysene equivalent	Sediments ($\mu g\,l^{-1}$ dw) chrysene equivalent	References
River mouth;			
Bangpakong	0.06–0.68		
Chaophraya	0.19–0.80	n.d.	Chartkittikunwong, 1986; Onodera et al.,1987
Thachin	0.26–0.55		
Bangpakong	0.11–0.25	0.44–0.52	
Chaophraya	0.21–0.43	0.72–1.57	Makjun, S. and Ponmanee, 2010
Thachin	0.15–0.34	0.86–1.18	
Maeklong	0.13–2.06	0.11–0.34	
Mangrove swamp:			
Laem Fapa,	6.67–10.17	42.3–236.0	
Samutprakarn Takuapa,			Wattayakorn, 1989
Phang-nga	2.00–6.20	5.0–9.5	

n.d.: no data

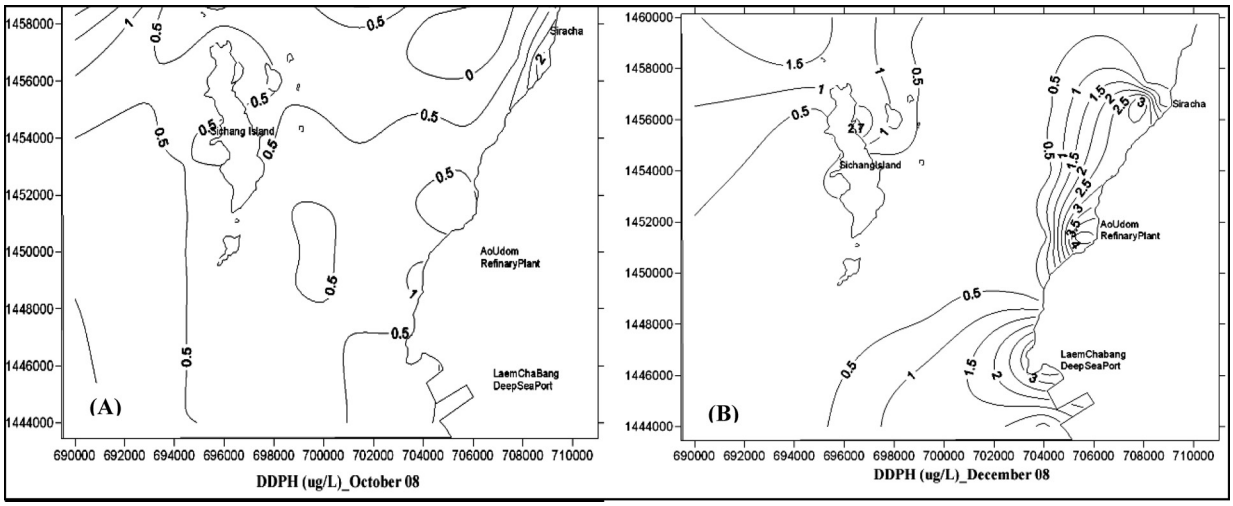

Fig. 3. Contour map of DDPH concentrations in Sriracha-Sichang coastal area in October (A) and December 2008 (B). After Wattayakorn and Rungsupa (2011).

Gulf of Thailand during 1994–1995 found that coastal waters of the inner Gulf and the eastern coast of the Upper Gulf exhibited occasional acute pollution events (DDPH $>40\,\mu g\,l^{-1}$ chrysene equivalents), superimposed 25% of the time upon chronic pollution ($\sim 4\,\mu g\,l^{-1}$) due to limited flushing of the inner Gulf and the eastern coast, and the presence of slightly contaminated water ($<1.2\,\mu g\,l^{-1}$) 75% of the time. Chronic petroleum hydrocarbon contamination in the nearshore waters was due to the consequence of low level discharges of oil from coastal shipping/boating activities, via urban, industrial, refinery and municipal wastes. Additional oil contamination could also originate from maritime transportation of crude and refined oil through the Thai waters, as a result of the discharge of ballast water from tankers. Increased pollution in the form of tarballs and oil slicks were observed in the past years (Wattayakorn et al. 1998). However, there was no significant increase of hydrocarbon levels in seawater of the Gulf of Thailand as a whole through the study periods, although the levels of hydrocarbon fluctuated throughout the year and are subjected to seasonal variations such as the monsoon seasons (Wattayakorn and Rungsupa 2011).

Petroleum hydrocarbon content in the sediments is probably a better measure of oil pollution. Generally petroleum hydrocarbons in water are partially degraded, with the residue accumulating in the bottom sediments. Hence, the hydrocarbons in the sediments were associated with heavier petroleum fractions which are easily adsorbed. Sediments containing fine particles tend to be good accumulators of organic pollutants presumably because of their greater effective surface area. Coarse-grained sands, on the other hand, can be

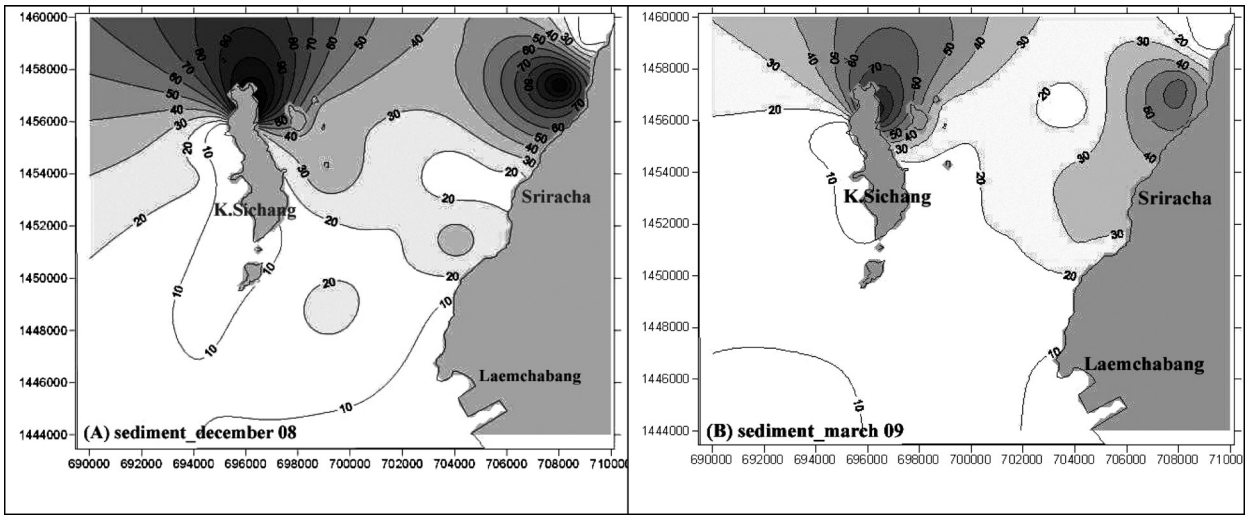

Fig. 4. Surface sediment contour map of petroleum hydrocarbons in Sriracha-Sichang coastal area. After Wattayakorn and Rungsupa (2011).

expected to have lower levels of hydrocarbon content, even though the corresponding levels in the seawater might be high. According to statistics of the Marine Department of Thailand (2009), there were more than fifty cases of oil spills from tanker/ship accidents and spillages along the Eastern Seaboard of the Upper Gulf during the last two decades. The high levels of hydrocarbons found in the inshore surface sediments of Sriracha and Sichang Island along the east coast of the Upper Gulf reaffirm anthropogenic inputs of oil into this coastal marine environment (Fig. 4). The level of hydrocarbons reported in the sediment samples ranged from 1.1 to 153.4 $\mu g\,g^{-1}$ dry weight, with the mean value of 29.4±30.0 $\mu g\,g^{-1}$. Concentrations of hydrocarbons in sediments of the inshore area (mean=40.9±39.9 $\mu g\,g^{-1}$) were significantly ($P<0.01$) higher when compared to the hydrocarbon concentrations of the offshore sediments (mean=23.3±16.3 $\mu g\,g^{-1}$) (Wattayakorn and Rungsupa 2011). Studies carried out by Suthanaruk (1991) and Tappatat (1995) showed that coastal sediment samples along the Eastern Seaboard of the Upper Gulf exhibited n-alkanes ranging from n-C_{15} to n-C_{33}. The patterns of n-alkane distributions and other indices , such as pristane/phytane, unresolved complex mixture (UCM) and carbon preference index (CPI) indicated that aliphatic hydrocarbon accumulation in the sediments were from both natural and anthropogenic origins.

Basically, higher petroleum hydrocarbon levels were found in the estuarine and nearshore sediments than in the offshore sediments; and the Upper Gulf sediments were more contaminated by petroleum hydrocarbons than the Lower Gulf area (Table 2). Apparently, some inshore sediment samples from the Gulf of Thailand showed petroleum hydrocarbon concentrations higher than 10 $\mu g\,g^{-1}$ and some stations showed concentrations even higher than 100 $\mu g\,g^{-1}$ (Table 2), and thus correspond to hydrocarbon concentrations in coastal areas considered low to moderately contaminated (NAS 1975, FAO 1982). Nevertheless, the levels of petroleum contaminants in surface sediments of the offshore water were generally comparable to the concentrations observed in relatively uncontaminated areas elsewhere in the world.

Polycyclic aromatic hydrocarbon contamination

Hydrocarbons with multiple ring structures are collectively referred to as polycyclic aromatic hydrocarbons, commonly abbreviated as PAHs. They are taken as an indicator of petroleum persistence since they are known to weather more slowly than aliphatic hydrocarbons and are associated with the toxicity of petroleum in the environment. PAHs are of considerable concern for the marine environment due to their relative persistence, carcinogenic and mutagenic potential as well as to their possible function as indicators of anthropogenic pollution. They are present, in greatly varying concentrations, in most marine environments. PAHs can arise in the environment from natural sources, oil and petroleum products and combustion processes. Although oil spills influence PAH concentrations in local areas, the major sources of PAH are anthropogenic and derived from land-based combustion sources. PAHs are globally distributed, and the highest concentrations generally occur close to urban centers. PAHs are relatively insoluble, adsorb strongly to particulate matter, and accumulate in bottom sediments (Blumer 1976, Laflamme and Hites 1978, Pashin and Bakhitova 1979, Quddus Khan et al. 2005, Zuo et al. 2007). More than one hundred PAHs have been found in nature; however, only sixteen have been selected as “priority pollutants” on the basis of frequency of occurrence and carcinogenicity as suggested by the United States Environmental Protection Agency (US EPA). These PAHs include naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenzo-

[a,h]anthracene, benzo[g,h,i]perylene, and indeno[1,2,3-c,d]pyrene. In analysis of these complex matrices, capillary gas chromatography (GC-FID) or combination of GC and mass spectrometer (GC/MS), rather than liquid chromatography (HPLC-FLD), is often the preferred approach for separation, identification, and quantification of PAHs, largely because GC generally affords greater selectivity, resolution, and sensitivity than HPLC. The combination of GC/MS can not only increase sensitivity and resolution, but also aid identification of PAHs in the sample. Generally lower detection limits (by an order of magnitude or more) were reported for GC/MS than for HPLC-FLD (Chiu et al. 1997, Gratz et al. 2000).

PAHs have been determined in coastal and marine environmental samples from the Gulf of Thailand since the early 1990s, but have not been routinely included in the national monitoring program due to sample preparation step is tedious, involving much time and effort, in order to obtain a clear extract for low-level PAHs analysis. Most PAH studies in Thailand were carried out in sediment samples from the estuarine and coastal areas, employing standard methodology for the isolation of PAHs from environmental samples, and subsequent analysis by GC-FID, GC/MS, HPLC-FLD and HPLC-DAD (as described in the references in Table 3).

An investigation of PAH contamination in sediments from the Thachin estuary by Wattayakorn and Chaipuriwong

Table 3. Ranges and mean concentrations of total polycyclic aromatic hydrocarbons in water and sediments from the Thai waters.

Area	Water (ng l^{-1})	Sediment ($\mu g\, g^{-1}$ dw)	Analytical methods	References
		Marine and coastal areas		
Map Ta Phut, Rayong (Ship-breaking area)	n.d.	0.003–0.32 (0.05±0.07)	GC-FID	Suthanaruk, 1991
Gulf of Thailand (offshore sediments)	n.d.	0.01–0.32 (0.05±0.03) Upper Gulf: 0.04–0.09 East coast: 0.01–0.32 Lower Gulf: 0.01–0.07	GC-FID	Sarin, 1994; Wattayakorn, 2003b
Coastal area of Rayong	n.d.	trace–1.99 (0.28±0.18)	GC-FID	Tappatat, 1995
East coast	n.d.	0.24–1.15	GC-FID	Duangkaew, 2003
Gulf of Thailand (coastal sediments)	n.d.	0.006–0.23 (0.05 ± 0.06)	GC/MS	Boonyatumanond et al., 2006; 2007a,b
		Rivers/Estuaries		
Chaophraya River	2.47	n.d.	GC/MS	Ehrhardt et al., 1990
Chaophraya River mouth	n.d.	0.60–10.7	GC-FID	Wiroonphol, 1990
Thachin estuary	n.d.	0.21–7.24 (2.60±1.99)	GC-FID	Sunwanich, 1991; Sunwanich & Wattayakorn, 1991
Chaophraya estuary	n.d.	0.58–4.71 (2.04±1.16)	GC-FID	Nokyoo, 1995
Thachin estuary	n.d.	0.26–1.52 (0.62±0.30)	GC-FID	Chaipuriwong, 2001; Wattayakorn & Chaipuriwong, 2006
Chaophraya & Thachin estuaries	192–1710* (693±447)	0.26–4.71** (1.53±1.73)	*HPLC-FLD **GC-FID	Wattayakorn, 2001; Wattayakorn, 2003a, b
Chaophraya River, estuary and canals	n.d.	0.03–2.51 Canals: 0.51–2.51 (2.29±2.56) River: 0.08–0.68 (0.26±0.17) Estuary: 0.03–0.72 (0.18±0.22)	GC/MS	Boonyatumanond et al., 2006; 2007a,b

n.d.: no data

(2006) provided useful information for evaluating PAH contamination levels in the Thai estuarine environment, as well as information on the possible sources of these PAHs. The total PAHs (∑PAHs) in sediments from the Thachin estuary were found to range from 0.26 to 1.52 $\mu g\,g^{-1}$) dry weight with a mean value of $0.62\pm0.30\,\mu g\,g^{-1}$). Dominant PAHs found were naphthalene, biphenyl, acenaphthene, fluorene, phenanthrene (Phe), chrysene, perylene, benzo (a) anthracene, anthracene (Ant), fluoranthene (Flt) and pyrene (Pyr). Distribution of PAH concentration in surface sediments showed high accumulation near the sources, with the predominance of unsubstituted PAHs in most samples, suggesting that most of the Thachin estuarine sediments exhibited pattern of pyrogenic inputs. PAH sources in these sediments were identified by Flt/Pyr, Phe/Ant, ∑MPhe/Phe and (2+3-ring)/(4+5-ring) PAHs ratios. It was found that the primary sources of PAHs in sediments were mostly of pyrolytic origins; such as from combustion processes through run-off, industrial and sewage discharges, and atmospheric input rather than spilled oils. It is important, however, to emphasize that the ratio of pyrolytic to petrogenic input of PAHs can be continuously changed because of the intensive industrialization and shipping activities in the Thachin estuary. In general, it can be concluded that the Thachin estuary was moderately contaminated as judged from PAH concentrations in the sediments.

A more intensive study regarding the status of PAH contamination in coastal and riverine environments in Thailand was carried out by Boonyatumanond et al. (2006, 2007a), where surface sediment samples were collected from canals, a river, an estuary, and coastal areas in Thailand; and sediment cores collected from the offshore transect of the Upper Gulf, then analyzed for PAHs by GC/MS. The ∑PAH concentrations ranged from 0.006 to 8.40 $\mu g\,g^{-1}$ dry weight. High emissions of PAHs from Bangkok city were evident by extremely high concentrations of PAHs in the urban canal sediments, ranging from 0.71 to 8.40 $\mu g\,g^{-1}$. In the Chaophraya estuary and the inner Gulf of Thailand, PAH concentrations clearly decreased offshore ($\sim0.05\,\mu g\,g^{-1}$). The average ∑PAH concentrations were $2.29\pm2.56\,\mu g\,g^{-1}$ in canals, $0.26\pm0.17\,\mu g\,g^{-1}$ in the river and $0.18\pm0.22\,\mu g\,g^{-1}$ in the estuary. Comparison of total PAH concentrations in estuarine and coastal sediments from Thailand to those in rivers, lakes and coastal zones of other Asian countries and other regions of the world, ranked PAH contamination in the Thai sediments as low to moderate (Fig. 5).

Source apportionment of sedimentary PAHs in the study area was carried out using the sum of methylphenanthrenes to phenanthrene (∑MP/P) ratio which indicated that sedimentary PAHs in the canals of Bangkok were of petrogenic origin. Further analysis with other oil fingerprinting biomarkers such as hopanes indicated that street dust was one of the major sources of petrogenic PAHs in the canals (Boonyatumanond et al. 2007b). On the other hand, low levels of PAHs with a pyrogenic signature were widely recorded in sediments from the remote areas of the coast and the Chaophraya River. The middle and lower reaches of the Chaophraya River, the river mouth, and the Upper Gulf of Thailand showed intermediate concentrations and profiles of PAHs indicating mixtures of petrogenic and pyrogenic origins. Sedimentary PAH concentrations normally decreased offshore with a half distance of about 10 km in the Upper Gulf off the mouth of the Chaophraya River (Boonyatumanond et al. 2006).

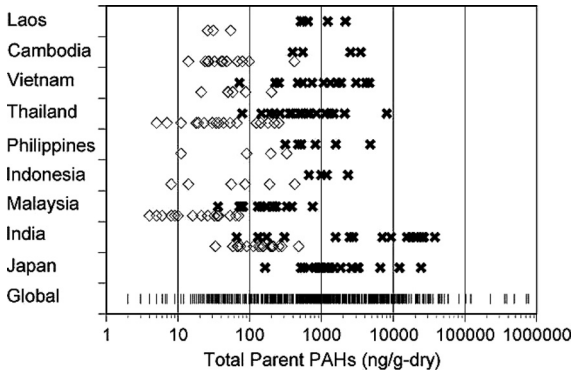

Fig. 5. Concentrations of total PAHs in sediments from Thailand in comparison to those in other tropical Asian countries, Tokyo, Japan and in world rivers, lakes and coastal zones. After Saha et al. (2009).

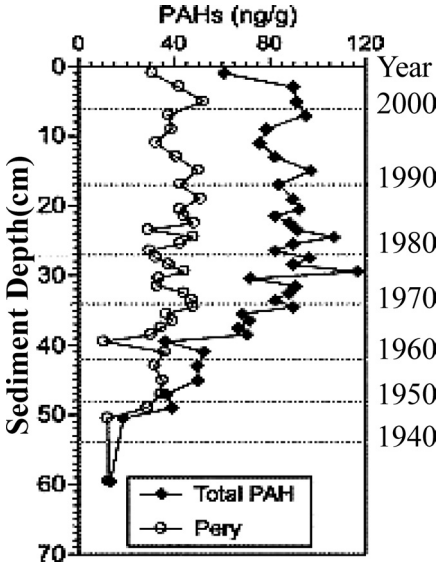

Fig. 6. Vertical profiles of total PAHs and perylene concentrations in a sediment core from the inner Gulf of Thailand. After Boonyatumanond et al. (2007a).

The historical record for PAHs found in the ^{137}Cs-dated sediment cores from the inner Gulf; collected at four locations along the transect line from the mouth of the Chaophraya River to offshore water, indicated that PAH concentrations increased sharply from the 1950s and 1960s (Fig. 6), with PAH compositions changed from a pyrogenic signa-

ture to a petrogenic signature. This was attributed to the increased industrialization and motorization in Bangkok during such time. PAH concentrations in the upper layers corresponding to the 1970s and later remained almost uniform or increased to the past 30 years, suggesting that the inputs of PAHs to Thailand coastal environment have not been decreasing during the past three decades, probably due to industrialization and urbanization in the catchment area. This contrasts with results in industrialized countries (i.e. Japan, USA), where PAHs decreased from the 1960s to the 1990s in coastal and lacustrine sediment cores (Boonyatumanond et al. 2007a).

These two studies and other data in Table 3 indicated that total PAH levels found in sediments from the Gulf of Thailand were not high when compared to other contaminated regions of the world, although some hot spots were found near urban centers, industrial sites, harbors and port/pier areas.

Conclusions

Oil in the sea from anthropogenic sources, whether from spills or chronic releases, is perceived as a major environmental problem in Thailand. From the studies reviewed, several analytical methods were used for determining hydrocarbon concentrations in seawater. The UVF method has been the most common and widely used in Thailand due to the advantage of being a rapid, uncomplicated screening method for monitoring petroleum hydrocarbon concentrations in seawater. However, because of limited spectral resolution, the UVF method is not suitable for determining the composition of the polyaromatic hydrocarbons. For sedimentary hydrocarbon analysis, both gas chromatography with flame-ionization detection (GC-FID) and GC/MS with computer-reconstructed mass fragmentograms were employed for the identification and quantification of the petroleum hydrocarbons. In addition to PAHs, the former technique also provides parameters such as the carbon-number preference index, which distinguishes biogenic from petrogenic n-alkanes, while the relative abundances of ions characteristic of triterpanes indicate similarities among samples, and give further evidence of the petrogenic origin of the hydrocarbon assemblages found (Butler and Sibbald 1987, Zakaria et al. 2001, 2002a, b, 2006, Boonyatumanond et al. 2007b).

The studies reviewed clearly showed that chronic petroleum hydrocarbon contamination is evident in the nearshore waters of the Gulf of Thailand, due to the consequence of low level discharges of oil from land-based sources and coastal shipping/boating activities. Additional oil contamination could also originate from maritime transportation of crude and refined oil through the Thai waters, as a result of the discharge of ballast water from tankers. Generally, higher levels of petroleum hydrocarbons were reported in estuarine and coastal waters of the inner Gulf than that in the offshore water of the Lower Gulf. Concentrations of hydrocarbons in sediment samples collected near major urban cities and industrialized locations are generally higher than those rural samples. No significant increase of hydrocarbon levels in seawater of the Gulf of Thailand was observed throughout the study periods.

The studies reviewed revealed that the dominant source of PAHs in the marine environment of Thailand is pyrogenic in origin, and only near the discharged points is a strong petrogenic PAH profile observed. There is an increasing trend of PAH contamination in sediments in the estuarine areas of the Upper Gulf. The elevated petroleum hydrocarbons and PAH levels were found in some samples from the canals and estuarine areas of the inner Gulf. The overall sedimentary PAH concentrations in the Thai waters, however were considerably lower than the Effects Range-Low (ER-L) of 4.02 $\mu g\, g^{-1}$ associated with potential adverse biological effects (Long et al. 1993). Nevertheless, management measures to minimize petroleum loading are needed to reduce long-term risk to the marine environment of Thailand. Additional data are also needed on the acute and chronic effects of sedimentary PAHs on benthic communities and other organisms, so that ecological risk assessment in coastal and estuarine environments can be achieved.

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