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Petroleum hydrocarbon residues in the marine environment of Koh Sichang-Sriracha, Thailand

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Abstract—This study describes petroleum hydrocarbon (PHC) contamination in water, sediment and green mussels (*Perna viridis*) of the coastal area of Koh Sichang-Sriracha, Chonburi along the Eastern coast of the Gulf of Thailand. The concentrations of PHC in coastal waters, as determined by UVF technique, were found to range from 0.01–12.55 $\mu g\,l^{-1}$ (chrysene equivalents) with higher values generally confined to port/pier areas. Analysis of surface sediments for PHC revealed that most of the sediments contained appreciable quantities of PHC. The concentrations of PHC in surface sediments varied from 1.1 to 153.4 $\mu g\,g^{-1}$ dry weight, with the mean value of 29.4±30.0 $\mu g\,g^{-1}$. The majority of values of PHC concentration in the surface sediments of Koh Sichang-Sriracha coastal area exceeded 10 $\mu g\,g^{-1}$ and the trend is indicative of transfer of PHC loads from the inshore areas to the offshore sediments. The average concentration of PHC burdens in the mussel tissues at Sriracha ranged from 8.1 to 161.0 $\mu g\,g^{-1}$ wet weight, with the average value of 43.7±55.5 $\mu g\,g^{-1}$, while that at Koh Sichang ranged 9.6–62.4 $\mu g\,g^{-1}$and the mean value was 23.1±17.2 $\mu g\,g^{-1}$. Bioconcentration factor (BCF) for PHC in green mussels was determined and found to vary from 1.7×10^4 to 3.5×10^5. The possible sources of oil contamination in the coastal waters of Koh Sichang-Sriracha marine area were the discharge of oily wastewater and lubricating oil from intensive shipping/boating activities at the port/pier areas, disposal of wastes from anchorage vessels, municipal and industrial discharges.

Key words: hydrocarbons, green mussel, *Perna viridis*, Koh Sichang, Thailand

Introduction

Petroleum-derived contaminants constitute one of the most prevalent sources of environmental degradation in the industrialized world. There are several sources of petroleum hydrocarbons, however, human activities such as oil transportation and spills, shipping, industrial, storm-water and domestic discharges are believed to be an important influence on hydrocarbons in the coastal marine environment. Petroleum hydrocarbons, especially the aromatic hydrocarbon fraction, are highly toxic to marine organisms at very low concentrations. In addition to direct lethal effects, petroleum hydrocarbons can exert sub-lethal effects, including reduced growth, altered feeding behavior, and lower reproductive success (GESAMP 1993, USEPA 1996, Quddus Khan et al. 2005).

The marine environment of Koh Sichang and Sriracha along the east coast of the inner Gulf of Thailand is considered an important resource for fishing, tourism, and recreation. Koh Sichang was once the summer residence of H.M. King Chulalongkorn, Rama V, during the nineteenth century. It is the nearest island to Bangkok, and as such is used as a retreat by many of the city's residents. The marine area of Koh Sichang and Sriracha supports numerous species of marine organisms, major commercial and recreational fisheries as well as reef coral habitats (Sakai et al. 1986, Yamazato and Yeemin, 1986, Kritsanapuntu et al. 2001a, 2001b, To-on 2003). Yet the area has also been assigned as a deep anchorage area for tankers, cargo ships and dozens of barges which transship their cargoes to lighter boats for the trip up the Chaophraya River to the Bangkok Port. Hence, this area is under considerable threat from intentional or accidental oil spills, ballast water discharged, as well as vulnerable to oil pollution due to oily water discharged from both anchored and moving vessels, and uncontrolled sewage and industrial wastewater discharges. Oil spills can cause damage to fishing and aquaculture resources by physical contamination, toxic effects and by disrupting business activity. According to statistics of the Marine Department of Thailand (2009), there were more than twenty cases of oil spills in these areas during the last two decades. Oil spilled in sheltered areas may persist in sediments for decades, where it can continue to exert negative effects on benthic organisms. Therefore, it is crucial to monitor the level of petroleum hydrocarbons in order to protect these coastal ecosystems.

This paper reports the findings of a comprehensive study conducted to assess the reference information on the levels of petroleum hydrocarbons contamination of water, surface sediments and mussels of the coastal environment of Koh Sichang and Sriracha in order to ensure the sustainable use of the marine environment.

Materials and Methods

Study area

Koh Sichang (Sichang Island) is a group of small islands located in the inner part of the eastern seaboard of the Gulf of Thailand (13°09′N, 100°49′E). It is relatively flat, consisting mainly of rock. The western coastline has a narrow rocky shore no greater than 100 m from the top of the beach to the edge of the intertidal area. All areas are exposed to air during low spring tides. Three types of climatological condition annually prevail in the Gulf of Thailand; southeast monsoon (Feb. to April), northeast monsoon (Oct. to Feb.) and southwest monsoon (May to Sep.). Tides are semi-diurnal with an average tidal range of 1.5 m. According to the morphology of the coastal area the main direction of the current runs more or less parallel to the shore line in a north-south direction. The flood current is from north to south and the mean rate is 0.2–0.5 m s^{-1}, the ebb current is from north to south and the mean rate is 0.1–0.3 m s^{-1}. The average annual precipitation is 1220 mm.

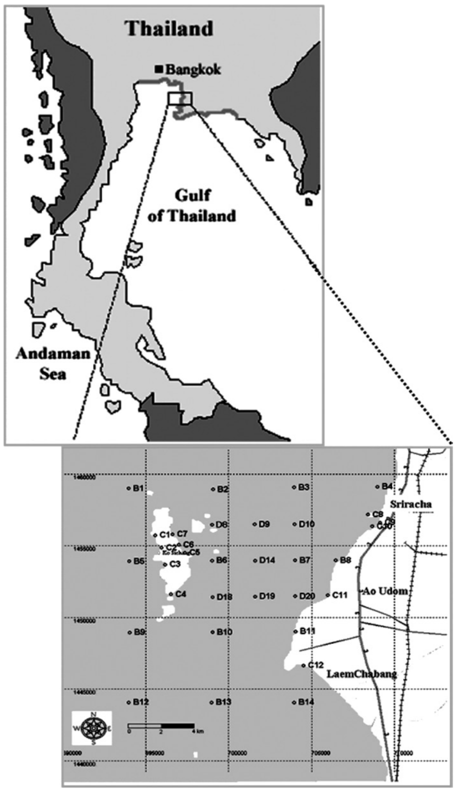

Fig. 1. Map of the marine area of Koh Sichang-Sriracha showing sampling locations.

Sampling

The locations of the sampling sites are shown in Fig. 1. Sampling took place between October 2008 and May 2009. Subsurface water samples (1 m deep) were collected using a 4-liter amber-glass bottle, which had been thoroughly cleaned with detergent, distilled water, acetone and hexane prior to use, held in a stainless-steel frame and operated by a handline. Samples of surface sediments were collected by a modified Van Veen grab sampler (0.1 m^2). Undisturbed top 5 cm sediments were sampled and placed in pre-cleaned glass jars and stored at −20°C until further analyses. Mussels (*Perna viridis*) of uniform size (7–8 cm in length) were obtained monthly from experimental mussel farms located in Sriracha Bay and Sichang Marine Science Training Station.

Analytical methods

Total dissolved/dispersed petroleum hydrocarbons (DDPH) in seawater samples were extracted using nano-grade n-hexane, evaporated using a rotary evaporator, taken up in 10.0 ml n-hexane into a vial and determined by UV fluorescence spectroscopy (UVF) on a Perkin–Elmer LS 50B spectrofluorometer. The fluorescence of the samples was measured at 310 nm excitation and at 360 nm emission wavelengths. Chrysene was used as the standard reference compound. Solvent blanks were run with each batch of samples (IOC/UNESCO 1984). The detection limit (DL=average ±3SD) was 0.05 $\mu g\,l^{-1}$ chrysene equivalents.

The petroleum hydrocarbon content in the sediments was estimated according to the method of IOC/UNEP/IAEA (1992). The frozen samples were dried in the freeze dryer, ground into powder and sieved using a 2 mm mesh sieve to remove coarse substances. Ten g of the dried sample was extracted with 150 ml of dichloromethane for 8 h in a soxhlet apparatus. The temperature for the soxhlet extraction was set at 40°C. After extraction, the solvent was evaporized and the residual oil was concentrated in 1 ml of n-hexane. After clean-up by silica gel column chromatography to remove biogenic lipid compounds, the petroleum hydrocarbon was then determined by UVF method as described for the water sample. Percentages of organic carbon in sediment samples were determined according to Schumacher (2002).

Mussel soft tissue was analyzed using procedures detailed previously (Burns and Smith 1981). Briefly, mussel tissues were soxhlet extracted with methanol and dichloromethane and the extracted hydrocarbons partitioned into 1 ml of n-hexane hexane. After clean-up by silica gel column chromatography to remove biogenic lipid compounds, the petroleum hydrocarbon was then determined by UVF method as described for the water sample.

Results and Discussion

Water Samples

The UVF method involves the excitation of electrons in the UV region of the electromagnetic spectrum and affects only those compounds with excitable electrons. Within a mixture of petroleum hydrocarbons, these are the polycyclic aromatic hydrocarbons (PAHs), whose harmful effects on living organisms are well known (GESAMP 1993, USEPA 1996). Unlike many aliphatic hydrocarbons, aromatic compounds 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). For these reasons, as well as reasons of simplicity, sensitivity, and speed, UVF has found general acceptance for the determination of petroleum hydrocarbons in environmental samples from the Gulf of Thailand and other regions.

The results of seawater analyses are given in Table 1. The range of DDPH in seawater around Koh Sichanh-Sriracha area found in the present study varied between 0.01 and 12.55 $\mu g\,l^{-1}$ chrysene equivalents. Inshore waters were found to have higher concentrations of DDPH than the offshore waters. The overall mean value of DDPH of the inshore waters was $1.65 \pm 2.14\,\mu g\,l^{-1}$ whereas that of the offshore waters was $0.40 \pm 0.29\,\mu g\,l^{-1}$.

The general pattern of DDPH distribution in seawater is shown in Fig. 2. As shown in Table 1 and Fig. 2, there are considerable variations in the concentrations of petroleum hydrocarbons in the studied area. This suggests that the distribution of oil is quite patchy and might reflect the intensity of shipping activities at the time of sampling. Higher concentrations were found in the inshore waters associated with port/pier areas. In general, the mean concentrations of DDPH in seawater were below the 5 $\mu g\,l^{-1}$ level (Coastal Water Standard set by the Pollution Control Department of Thailand) and ranged from 0.06 to 12.55 $\mu g\,l^{-1}$. Among all stations, Stns C7, C8 (near fishing piers), C12 (near a deep seaport) and C9 (near the wastewater discharge point of Sriracha Town) showed higher values than 5 $\mu g\,l^{-1}$ in some occasions during the period of investigation. The results also indicated that the inshore waters along Sriracha coast exhibited a higher mean concentration of DDPH ($2.24 \pm 1.53\,\mu g\,l^{-1}$) than that of Koh Sichang ($1.23 \pm 0.54\,\mu g\,l^{-1}$). According to Wattayakorn et al. (1989), the background value of DDPH for the inner Gulf of Thailand was estimated to be 1.2 $\mu g\,l^{-1}$ chrysene equivalents; evidently chronic DDPH contamination of the inshore waters of Sriracha coast has shown to be persistent in the area, with occasional acute pollution events at some sites. The DDPH concentrations found in the present study are comparable to the values previously reported for the Gulf of Thailand by Wattayakorn et al. (1998), Kan-atireklap et al. (2002), Kan-atireklap et al. (2005), Meepoka (2007) and Chomchuen (2008), suggesting the background ranges found at localities near to regular shipping and boating activities which, however, are much lower when compared to values reported during the oil spill incident at Laemchabang area in 2006 (48.07 $\mu g\,l^{-1}$) (Kan-atireklap et al. 2007) or near the main shipping lane in the Straits of Malacca (96–135 $\mu g\,l^{-1}$ chrysene equivalents) (Abdullah et al. 1996).

The results of petroleum hydrocarbons (PHC) for sediment analyses are given in Table 2. The general pattern of PHC distribution in surface sediments is shown in Figs 3 and 4. PHC recorded for inshore sediments fell within the range of 1.06 to 153.4 $\mu g\,g^{-1}$ chrysene equivalents, with the overall mean value of $41.1 \pm 43.1\,\mu g\,g^{-1}$ which was higher than that for the offshore sediments (mean value of $23.2 \pm 16.3\,\mu g\,g^{-1}$ and ranged from 2.63 to 78.6 $\mu g\,g^{-1}$).

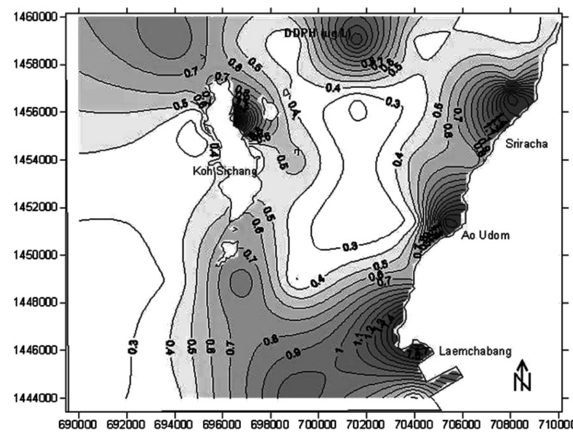

Fig. 2. Contour map of mean DDPH concentrations in the studied area.

Table 1. Mean concentrations of dissolved/dispersed petroleum hydrocarbons (DDPH) in seawater. Ranges are given in parenthesis.

Waters	DDPH ($\mu g\,l^{-1}$) in chrysene equivalents					
	Oct-08	Nov-08	Dec-08	Mar-09	Apr-09	May-09
Offshore	0.57±0.27 (0.29–1.33)	0.33±0.13 (0.17–0.57)	0.43±0.44 (0.14–1.94)	0.30±0.16 (0.13–0.71)	0.40±0.30 (0.06–0.99)	0.36±0.28 (0.06–1.05)
Inshore	0.76±0.59 0.10–2.26	1.21±1.59 0.20–5.90	1.70±1.77 0.13–4.81	1.63±2.4 10.18–9.05	1.29±1.14 0.28–3.56	3.33±3.46 0.50–12.55

Table 2. Mean concentrations of PHC and percentages of organic carbon (%OC) in surface sediments. Ranges are given in parenthesis.

Sediments	Dec-08		Mar-09	
	PHC ($\mu g\,g^{-1}$)	%OC	PHC ($\mu g\,g^{-1}$)	%OC
Offshore	25.2±19.3 (3.34–78.6)	1.30±0.27 (0.61–1.85)	21.22±12.7 (2.63–52.8)	1.43±0.18 (0.95–1.86)
Inshore	45.0±52.1 (1.06–153.4)	0.59±0.70 (0.15–2.15)	40.1±33.5 (4.57–107.6)	1.05±1.07 (0.06–2.73)

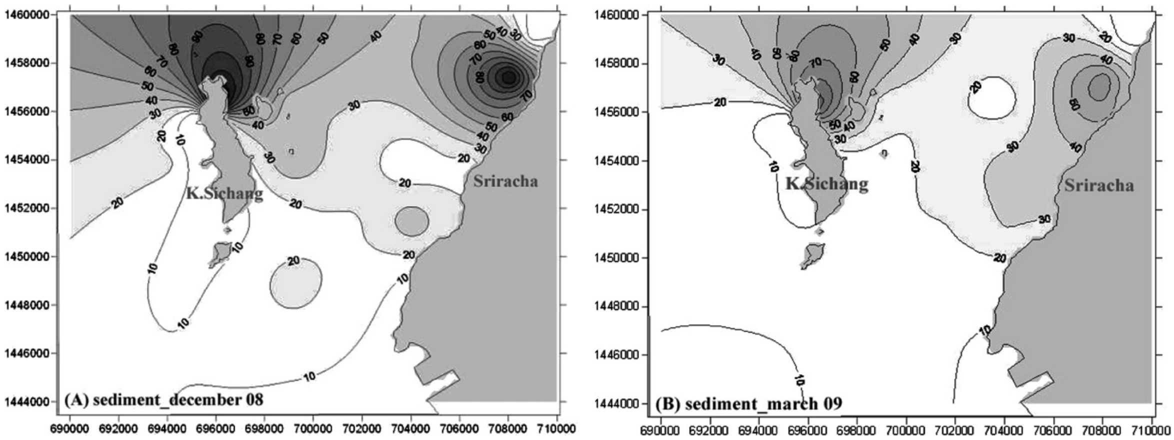

Fig. 3. PHC contour maps for sediments during the two sampling periods.

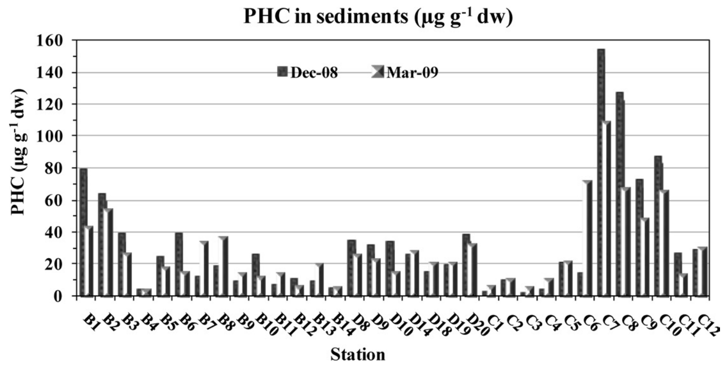

Fig. 4. PHC variation in surface sediments during the two sampling periods.

The PHC results for the sediments (Table 2 and Fig. 4) revealed that all of the sediments analyzed contained appreciable quantities of petroleum hydrocarbons. Intertidal and subtidal sediments are long-term repositories for hydrocarbons released into the environment, and expectedly, high concentrations of petroleum hydrocarbons are found in sediments of estuaries, harbours, bays and in coastal areas receiving industrial and urban discharges (Fowler et al. 1993, Abu-Hilal et al. 1994). Concentrations of PHC 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 PHC concentrations of the offshore sediments (mean=23.3±16.3 $\mu g\,g^{-1}$). Along the Sriracha coast, Stn C8 located at the pier area exhibited the highest PHC mean concentration of 96.1 $\mu g\,g^{-1}$, while along Koh Sichang coast, Stn C7 (also located at the pier area) showed the highest PHC concentration with the mean value of 130.6 $\mu g\,g^{-1}$. The highest concentrations indicate the influence of shipping/oil activities carried out in the two areas. In general, the inshore sediments along the Sriracha coast exhibited a higher mean PHC value (55.7±34.5 $\mu g\,g^{-1}$) than that of the Koh Sichang inshore sediments (30.7±46.6 $\mu g\,g^{-1}$). Comparing the concentrations of petroleum hydrocarbons found in the present study with the values reported for the Gulf of Thailand and those of selected marine areas (Table 3), it is clear that the present PHC concentrations were higher than those for Map Ta Phut and Ban Phe (Rayong), Sattahip and Angsila (Chonburi); and were comparable to those detected in coastal sediments of Straits of Johor (Malaysia) and Dabhol–Ratnagiri coast (India), but were much lower than those at the Trinidad coast. Apparently,

Table 3. Concentrations of petroleum hydrocarbons in sediments from the Gulf of Thailand and other similar marine environment (determined by applying fluorescence technique and using chrysene as a standard)

Study areas	Sediment (μg/g dw)	References
Map Ta Phut, Rayong (Ship-breaking area)	0.06–1.86 (0.50±0.36)	Suthanaruk, 1991
Ban Phe, Rayong	1.53±2.39	Sanguansin et al., 2003
Sattahip, Chonburi	0.39–12.98 (5.69±3.55)	Intang , 2006
Bansean & Angsila, Chonburi	0.37–23.55 (6.82±8.22)	Meepoka, 2007
Trinidad coast	3.0–1824.2	Agard et al., 1988
Straits of Johor, Malaysia	0.71–36.7	Abdullah et al., 1996
Dabhol-Ratnagiri coast, India	0.9–107.7	Chouksey et al., 2004
Koh Sichang & Sriracha, Chonburi	1.10–153.4 (29.4±20.0)	This study

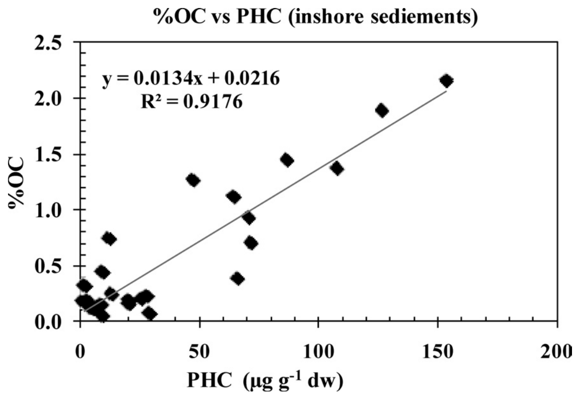

Fig. 5. The relation between concentrations of petroleum hydrocarbons and percentages of organic carbon (%OC) in the inshore sediments.

many inshore stations, particularly along the Sriracha coast, showed PHC concentrations much higher than 10 μg g^{-1} and some stations (i.e. Stns C7 and C8) showed PHC concentrations even higher than 100 μg g^{-1} and thus correspond to hydrocarbon concentrations in coastal areas considered low to moderately polluted (NAS, 1975; FAO, 1982).

The relationship between concentrations of PHC and percentages of organic carbon (%OC) in the inshore sediments (Fig. 5) indicates a very good correlation between the two components and implies that the increase in petroleum hydrocarbon contamination would result in an increase in organic carbon contents. However, no significant correlation was found between the percentage of organic carbon and PHC concentrations in the offshore sediments ($P>0.05$) (graph not shown). This probably is due to a rather high organic matter content from spills of raw materials such as fertilizer, cassava (tapioca) and grains from ships/barges during the loading and unloading activities around the anchorage area which were then added into the organic content of offshore sediments in addition to waste water discharges and oil spills from ships. The composition of petroleum-hydrocarbon mixtures also changes owing to different sources and physical, chemical, and biological influences. For this reason, results originating from various areas, even those obtained under identical conditions, are only conditionally comparable.

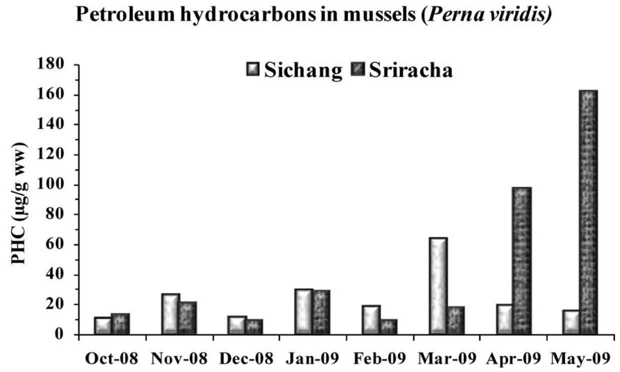

Fig. 6. Accumulation of PHC in mussel (*Perna viridis*) tissues from the two experimental farms during the studied period.

Mussels

The results for mussels, expressed on wet weight concentration basis are given in Fig. 6. Mussels are known to accumulate trace contaminants such as heavy metals and hydrocarbons over time and concentrate minute amounts of these contaminants present in the water column. The pattern of PHC accumulation at both sites seems to mimic the DDPH concentrations of the surrounding seawater. The average concentration of PHC in mussel tissues at Sriracha, during October 2008 to May 2009 study period, was 43.7±55.5 μg g^{-1} which was higher than that at Koh Sichang (mean 23.1±17.2 μg g^{-1}), and was in good agreement with the concentration levels of DDPH found in the two studied areas.

The potential of a substance to bioaccumulate in an organism can be expressed by the bioconcentration factor (BCF). The BCF measures the concentration of a particular substance in a living organism relative to its concentration in the surrounding medium. Calculation of BCF assumes that the mussels were in equilibrium with the hydrocarbon levels found by the samplers. That is, it is assumed that the mussels were not reflecting higher or lower concentrations that may have existed before the samplers were deployed. The BCF for PHC in green mussels were calculated and found to vary from 1.7×10^4 to 3.5×10^5 (mean 3.1×10^4) at Sriracha, while that at Sichang varied from 1.21×10^4–2.97×10^5 (mean 2.77×10^4). The BCF values were much higher than 5000, indicating a propensity of PHC to accumulate in lipids. The criteria to determine the propensity of an organic chemical to undergo bioaccumulation were specified in the Toxic Substance Management Policy (TSMP) under the Canadian Environmental Protection Act of 1999 (www.ec.gc.ca). However, it is very difficult to compare BCF with previously published values elsewhere because of differences in exposure conditions, experimental methodology and reporting format.

Conclusions

Petroleum pollution problems in the coastal waters of Koh Sichang-Sriracha marine area appear to arise from intermittent discharges to the marine area of crude oil, petroleum-refined products, ballast water and land-based wastewaters. Sediments act as pollutant sinks and provide an integrated picture of the events taking place in the water column; hence sediments are an extremely good indicator of the magnitude of environmental contamination resulting from an event such as an oil spill. Major oil spill incidents in the past represent a significant source of hydrocarbons locally and episodically. The present study presents a baseline distribution assessment of total hydrocarbons in the water, mussels and benthic sediments of Koh Sichang-Sriracha coastal and marine areas; from the results of this work, it is evident that all sites are contaminated to some extent with petroleum hydrocarbons. The overall levels of anthropogenic hydrocarbons in the surface sediments when compared to selected marine areas with substantial industrial and domestic coastal activities elsewhere, revealed a low to moderate level of hydrocarbon pollution. However, owing to PHC bioaccumulative potential and toxicity to both aquatic organisms (biota) and human consumption of seafood, as well as impacting the composition and diversity of infaunal communities, periodical monitoring and assessment of water, sediments and tissues of various biota of Koh Sichang and Sriracha coastal waters is still necessary.

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