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Petroleum hydrocarbon concentrations in eight mollusc species along Tamilnadu coast, Bay of Bengal, India

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Abstract

Eight mollusc species and sediment samples collected from three different stations along Tamilnadu coast, Bay of Bengal, India were analysed for the levels of petroleum hydrocarbons to elucidate the status of the petroleum residues in mollusc meant for human consumption. The concentrations of petroleum hydrocarbons in sediments along Tamilnadu coast varied from 5.04–25.5 μg/g dw (dry weight). High concentration of petroleum hydrocarbons in the sediment of Uppanar estuary (25.5 ± 1.45 μg/g dw) was perhaps land and marine based anthropogenic sources of this region. The petroleum hydrocarbon residues in eight mollusc species collected from Uppanar, Vellar and Coleroon estuaries varied between 2.44–6.04 μg/g ww (wet weight). Although the concentration of petroleum hydrocarbons in sediment of the Uppanar region was markedly higher than the background, the petroleum hydrocarbon residues in mollusc collected from Uppanar estuary did not suggest bioaccumulation. The results signified that industrial growth has affected the aquatic environments and regular monitoring will help to adopt stringent pollution control measures for better management of the aquatic region.

Key words: petroleum hydrocarbons; sediment; mollusc; pollution; Tamilnadu coast

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Introduction

Oil pollution can have a serious economic impact on coastal activities and on those who exploit the resources of the sea. In most cases such damage is temporary and is caused primarily by the physical properties of oil creating nuisance and hazardous conditions. The impact on marine life is compounded by toxicity and tainting effects resulting from the chemical composition of oil, as well as by the diversity and variability of biological systems and their sensitivity to oil pollution. Petroleum hydrocarbons enter into marine environment from a number of sources including industrial discharges, accidental spills, shipping activities, atmospheric fallout, and oil and gas exploration (NRC, 2003). The marine environment especially the near shore and estuarine areas are subjected to excessive contaminations by human activities. These vulnerable and ecologically sensitive areas are often selected for the establishment of major petroleum based industries for easy transportation and disposal.

GESAMP (1993) estimated a total input of oils at 2.3 million tonnes per year and ranked the sources as follows: land based sources (urban runoff, coastal refineries) 50%; oil transporting and shipping (operational discharges, tanker accidents) 24%; offshore production discharges 2%; atmospheric fallout 13%; natural seeps 11%. NRC (2003) reported that the average total worldwide annual release of petroleum from all known sources to the sea has been estimated at 1.3 million tonnes. According to the report, the main categories of sources contribute to the total input as follows: natural seeps 46%; discharges from consumption of oils (operational discharges from ships and discharges from land based sources) 37%; accidental spills from ships 12%; extraction of oil 3%.

The extent of contamination can be assessed by measuring pollutant concentrations in water, sediments and marine organisms. Although easier to process, water samples are difficult to interpret since the water is constantly flowing, transporting pollutants from one place to another while diluting them, often to concentrations below detection limits. Sediment samples, on the other hand, integrate chemicals over time and over the water column via precipitation, and organisms often concentrate pollutants in their tissues, facilitating the detection of specific substances. Marine organisms can be widely used as bioindicators and integrators of environmental conditions in marine environment. Determination of petroleum hydrocarbons

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in marine organism tissues is necessary because of their carcinogenic and mutagenic properties (Albers, 1995; Noreña-Barroso et al., 1999). Mussels have been considered as pollution sentinels by many researchers (Hellow et al., 2002) because of their bioaccumulation capacity, and the National Status and Trends Program (Wade et al., 1998) recommended them as a foreground matrix to environmental monitoring. Organisms such as shellfish have commonly been used to monitor pollution in marine waters due to their capacity to concentrate and retain persistent chemicals (Connell et al., 1999). Sediments are also ideal matrices for chemical analysis, as they are the final fate for a variety of lipophilic pollutants (Hellow et al., 2002), and reflect long-term deposition. Petroleum hydrocarbons enter the marine environment is removed by evaporation; a portion gets distributed in water, accumulated in sediment and transferred to biota (Chouksey et al., 2004).

The environmental chemistry of minor river basins in India has received less attention, despite some environmental studies in major rivers (Subramanian et al., 1987; Ramesh et al., 1989). Estuaries represent habitats at risk, receiving anthropogenic effluents from various rivers and waters, translocated from remote and nearby domestic, agricultural, and industrial sources of pollution (Wang et al., 2008; Zhang et al., 2008, 2009; Hui et al., 2009). India has a long coastline of 8129 km and of this 6000 km is rich in estuaries, creeks, brackish water, lagoons and lakes. The southeast coast of India is an important stretch of coastline with many significant landmark features, where many major rivers drain into the Bay of Bengal and they are also richer in marine fauna than the western coast of India.

The present study focuses on the level of petroleum hydrocarbons in sediments and eight mollusc species along Tamilnadu coast, southeast coast of India. The results from the present study will be useful for pollution monitoring programs along rivers and in coastal regions in India.

1 Materials and methods

1.1 Study area

Tamilnadu is the southern state on the east coast in India with a coastline of approximately 1000 km. This coast has extensive areas of estuaries, mangroves, brackish water lagoons all of which are connected to the Bay of Bengal as various locations along the east coast (Fig. 1). Cuddalore town is a significant coastal city in Tamilnadu State that hosts a number of large-scale industries. The Uppanar River runs parallel to the coast south of Cuddalore town, and a number of small streams of domestic effluents, and treated and untreated effluents from industries connect to the coast via the river. The main industries along the western bank of Uppanar River include chemicals, beverage manufacturing, tanneries, oil, soap, paint production, paper, and metal processing plants; all industries are located along its western bank. Cuddalore harbour located in the estuarine region of Uppanar River is occupied by a fleet of mechanized fishing boats, which operates in the

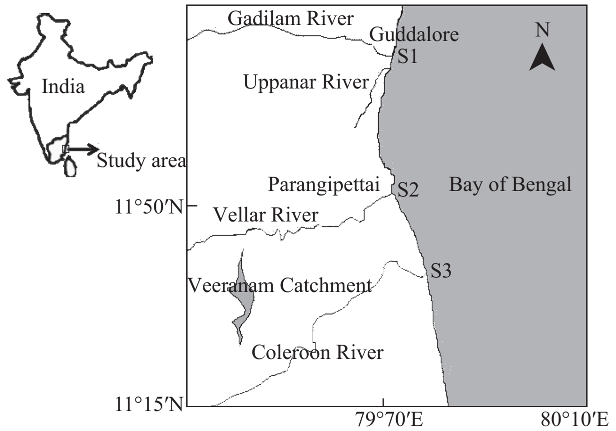

Fig. 1 Map of the study area showing the sampling sites along Tamilnadu coast.

coastal zone. The Vellar river originates at the Kalvarayan hills in the Salem district, runs for 90 km and flows through the borders of Villupuram and Perambalur districts for very few kilometers. In its stretch, it enters the Cuddalore district, flowing for another 105 km, and ends its journey into the Bay of Bengal at Parangipettai. The Coleroon estuary is located in southern part of the study area.

1.2 Sampling methods and petroleum hydrocarbon analysis

Eight different species of molluscs including *Saccostrea cucullata, Crassostrea madrasensis, Meretrix meretrix, Perna viridis, Meretrix casta, Telescopium telescopium, Anadara granosa* and *Hemifusus cochlidium* and sediment samples were collected from three stations (Uppanar (S1), Vellar (S2) and Coleroon (S3) estuaries) along Tamilnadu coast, Bay of Bengal, India during March 2010. There are ten samples from each kind of mollusc species. Sediment samples were collected from all the three sampling stations. Sediment samples were collected using a plastic spoon. The collected sediment was thawed, saponified using a KOH methyl alcohol mixture followed by extraction with *n*-hexane. The concentrated extract, after drying, was separated into alkane and aromatic fractions in an alumina column and the intensity of fluorescence of the aromatic fraction was measured (IOC-UNESCO, 1982).

The mollusc species were collected from the study areas by hand picking. The soft tissue was removed from the shells with a plastic knife and wrapped in aluminium foil, sealed in thick polythene bag and preserved at –20°C until analyzed (FAO, 1983). The thawed tissue was homogenized, saponified using KOH–methyl alcohol mixture, centrifuged and filtered. The filtrate was extracted with *n*-hexane, organic layer washed with distilled water, dried, evaporated to a small volume, chromatographed on alumina and fluorescence measured (IOC-UNESCO, 1984). Petroleum hydrocarbon concentrations in mollusc species were determined using UV-Fluorescence Spectroscopy (Varian Cary Eclipse, USA). The fluorescence of the samples was measured at an emission wavelength of 364 nm (excitation wavelength 310 nm). All blanks, standards and samples were measured in a Teflon capped 1 cm silica fluorescence cell under identical instrumental settings and conditions. Duplicates, spikes and blanks were treated identically using Chrysene as a standard reference to test the precision, accuracy and solvent purity in the analytical procedure and the data were expressed in terms of Chrysene equivalents. Percentage recovery for spiked samples ranged from 96% to 99%, whereas precision agreed within 5%. Blank values were almost negligible. All experiments were conducted in 5 replicates and the averages of the values were reported along with standard deviations. The results were treated statistically using the Student's *t*-test ($p < 0.05$).

2 Results and discussion

2.1 Sediment

The texture classes of sediments in Uppanar, Vellar and Coleroon estuaries are silty clay, silty sand and silty sand respectively. The higher mud (silt + clay) content in Uppanar (S1) is due to the low fluvial discharge and a better mixing of saline and fresh water that facilitated flocculation and faster settling of suspended particles (Nair et al., 1993). The high contents of silty sand in Vellar (S2) and Coleroon (S3) estuaries are due to the narrow mouth and the high energy waves, which do not allow free flow of fresh water into the coastal zone area. The fine sediment particles dispersed in the water column have high absorbing character and effectively scavenge the trace contaminants entering the marine area (Sahu and Bhosale, 1991).

The distribution of petroleum hydrocarbons (PHC) in sediments along Tamilnadu coast is shown in Fig. 2. The concentrations of PHC in sediments along Tamilnadu coast varies in a wide range of 5.04–25.5 μg/g dw (dry weight). The high concentration of petroleum hydrocarbons (25.5 ± 1.45 μg/g dw) was obtained at Uppanar estuary. The PHC value for Coleroon estuary is 5.55 ± 1.05 μg/g dw. The lowest PHC concentration 5.04 ± 1.00 μg/g found at Vellar (S2) is considered as background level for the present study. Jonathan et al. (2008) have found the low level of dissolved oxygen (DO) in Uppanar river waters. The low DO content occurred when industrial effluents pollute the waters and high chemical oxygen demand of the effluents resulted in reduced DO which is situated very close to the river bank, where the combined effect of temperature, photosynthetic action, and biochemical degradation of waters enters the water column (Ghosh et al., 1991). Sediments containing fine particles tend to be good accumulation of organic pollutants presumably because of their greater effective surface area (Venkatachalapathy et al., 2010a). The highest level of PHC in Uppanar estuary sediment might be the land based sources such as industrial effluents from the adjoining industries, municipal wastes, and atmospheric fallout and also the marine based sources such as, Cuddalore harbour and fishing activities. But in Vellar and Coleroon estuarine sediments, the PHC levels are nearly 5 times lower than the Uppanar estuary sediments. In the Bay of Bengal region, no natural oil seeps have been reported and accidental spills from ships are infrequent. Therefore, it may be inferred that discharges from ships and land based sources contribute more to the present level of PHC in this region. The overall petroleum hydrocarbon concentration in the Tamilnadu coastal sediments, although higher than the background, do not indicate overall levels of sediment contamination compared with values reported for selected costal areas around the world (Table 1).

2.2 Mollusc tissue

The petroleum hydrocarbon residues in mollusc species collected from Uppanar, Vellar and Coleroon estuaries are shown in Fig. 2. The PHC values varied between 2.44 and 6.04 μg/g ww (wet weight). The highest PHC values were obtained in Uppanar estuary mollusc samples. Concentrations of PHC in Uppanar estuary mollusc species decreased in order of *Crassostrea madrasensis* (6.04 μg/L) > *Saccostrea cucullata* (5.22 μg/L) > *Hemifusus cochlidium* (5.1 μg/L) > *Telescopium telescopium* (4.9 μg/L) > *Meretrix meretrix* (4.6 μg/L) > *Perna viridis* (4.1 μg/L) > *Anadara granosa* (3.95 μg/L) > *Meretrix casta* > (3.9 μg/L). Concentrations of PHC in Vellar estuary mollusc species decreased in order of *C. madrasensis* (4.55 μg/L) > *S. cucullata* (4.34 μg/L) > *H. cochlidium* (3.82 μg/L) > *P. viridis* (3.18 μg/L) > *T. telescopium* (3.07 μg/L) > *M. meretrix* (2.87 μg/L) > *M. casta* > (2.62 μg/L) > *A. granosa* (2.44 μg/L). Concentrations of PHC in Coleroon estuary mollusc species decreased in order of *C. madrasensis* (4.62 μg/L) > *S. cucullata* (4.21 μg/L) > *H. cochlidium* (3.90 μg/L) > *T. telescopium* (3.25 μg/L) > *P. viridis* (3.20 μg/L) > *M. meretrix* (3.01 μg/L) > *M. casta* > (2.95 μg/L) > *A. granosa* (2.60 μg/L). Mostly, the lowest petroleum hydrocarbon concentrations found in Vellar (S2) estuarine mollusc species are considered as background level for the present study. Among the three stations, the concentrations of PHC in all mollusc species are high in Uppanar estuary. This higher concentration might be caused by discharges of industrial and domestic effluents along the coast, boating and fishing activities from Cuddalore harbour. Among the eight mollusc species, *C. madrasensis* has highest PHC concentrations from all the locations due to high biotransformation of pollutants, which showed that *C. madrasensis* is good bio-indicator for PHC pollution along the coast. Commonly PHC values are found in the range of 0.1–10 μg/g (wet weight) in marine fish, while, fish from polluted areas can have PHC concentrations of 10–1000 times higher (Law et al., 1997; Chouksey et al., 2004).

Many publications suggested that pattern of distribution for lipophilic chemicals in aquatic system is transfer from low level tropic biota to higher level tropic biota to aquatic birds and marine mammal (Albers, 1995; Connell, 1997). However, bioavailability and organic physiology are the two important variables that have major effect on contaminant body burden. Physiological factors including lipid levels and the rates of uptake and elimination (metabolism, diffusion and excretion) also affect contaminant body burden (Connell, 1997). According to Massoud et al. (1996), petroleum hydrocarbon pollution levels in sediments < 15

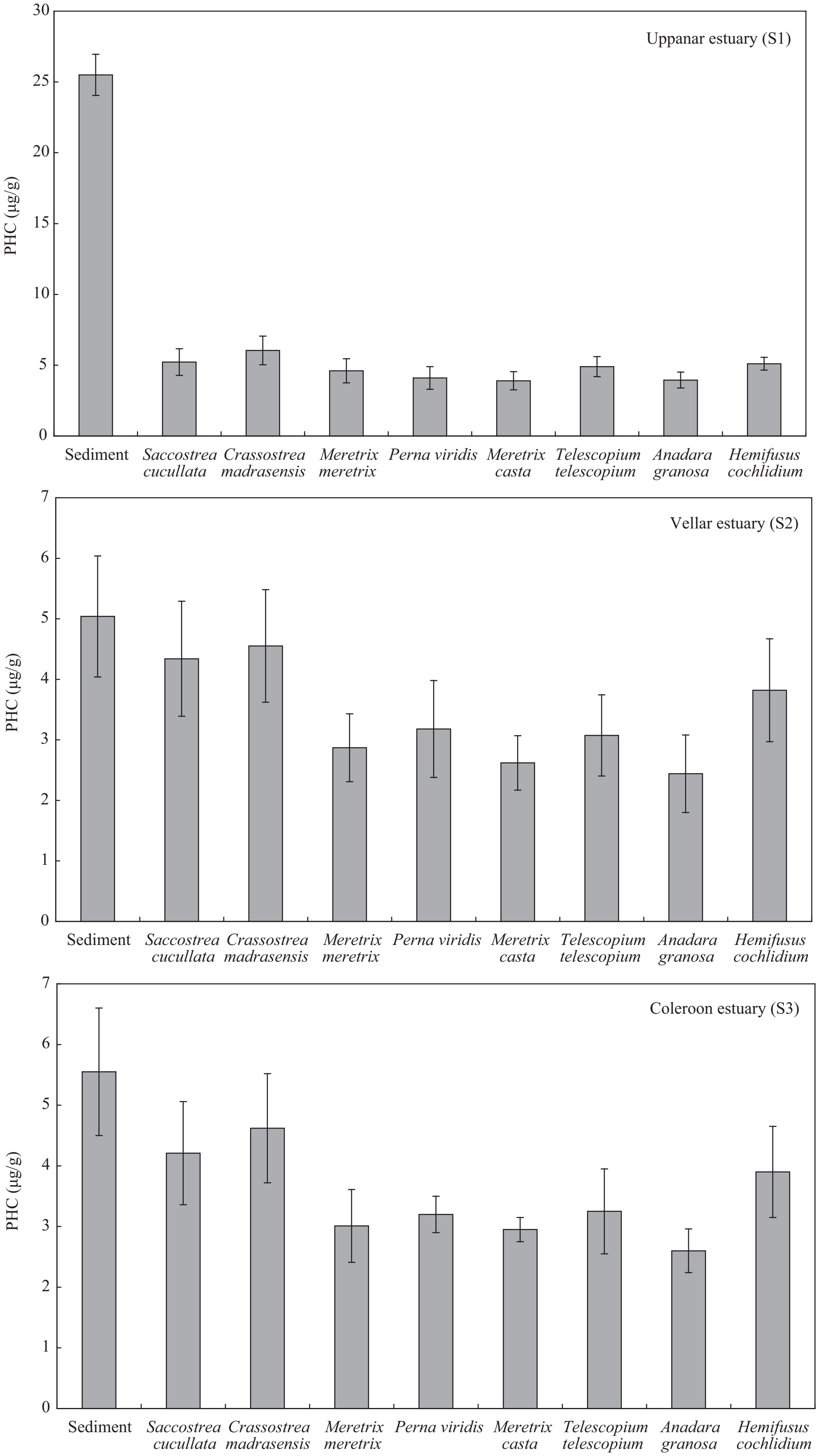

Fig. 2 Distribution of petroleum hydrocarbon (PHC) concentrations in sediments and mollusc species in Uppanar estuary, Vellar estuary, and Coleroon estuary. Data are presented as mean ± SD at $p < 0.05$.

Table 1 Comparison of petroleum hydrocarbon concentration in sediment along Tamilnadu coast with those of selected marine areas

Location	PHC (μg/g)	Source	Location	PHC (μg/g)	Source
Tamilnadu coast, India	5.04–25.5	Present study	Changjiang Estuary, China	2.2–11.82	Bouloubassi et al., 2001
Chennai coast, India	1.88–39.7	Venkatachalapathy et al., 2010b	Patos Lagoon Estuary, Brazil	39–11,780	Zanardi et al.,1999
Qua Iboe Estuary, Nigeria	18.01–210.23	Benson et al., 2008	Straits of Johor, Malaysia	0.7–36.7	Abdullah et al., 1996
Gulf of Fos, France	7.8–180	Mille et al., 2007	Tampa Bay, Florida, USA	200–4300	Sherblom et al., 1995
Guanabara Bay, Brazil	77–7751	Da Silva et al., 2007	Shetland Island, UK	7–8816	Kingston et al., 1995
Bay of Fort de, France	54–1045	Mille et al., 2006	Sao Sebastiao, Brazil	20–200	Medeiros and Bicego, 2004
Bizerte lagoon, Tunisia	0.05–19.5	Mzoughi et al., 2005	Liverpool Bay, UK	29.0	Law, 1981
Thane Creek, Mumbai coast, India	7.6–42.8	Chouksey et al., 2004	Scotian shelf, Canada	1.0–94.0	Keizer et al., 1978
Todos os Santos Bay, Brazil	8–4163	Venturini and Tommasi, 2004			

μg/g as Chrysene equivalents are considered to be the natural background level in this region. Hence, although the concentration of PHC in sediment of the Uppanar region was markedly higher than the background, there is no evidence for its increase in mollusc of the region. Moreover, further work is needed to indicate potential explanations for this observation.

3 Conclusions

The present study presents a baseline distribution assessment of petroleum hydrocarbons in coastal sediments and mollusc species along Tamilnadu coast, Bay of Bengal, India. The highest value of PHC obtained at Uppanar estuary might originated from land and marine based anthropogenic sources. Petroleum hydrocarbons have highly adhered to fine-grained particulates. Thus the sediment form a major sinks for organic matter. Even though the concentration of PHC in sediment of the Uppanar region was markedly higher than the background, there is no evidence for its increase in mollusc of the region. From a public health point, petroleum hydrocarbon residue levels in all mollusc samples analysed in this study were considerably lower than the hazardous levels. Presently, as Tamilnadu coastal area is in a rapid development stage of new harbour, chemical industries, power plants, oil exploration and other large scale industries, further assessment of petroleum hydrocarbons and the various hydrodynamic conditions acting in the region are needed to be conducted in detail. The results will be useful for pollution monitoring programs along rivers, and coastal regions which will also check the levels of petroleum hydrocarbons.

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