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ORIGINAL RESEARCH ARTICLE

Petroleum Hydrocarbons (PHC) in Sediments of Three Different Ecosystems from Southeast Coast of India

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ABSTRACT

The seasonal variation of petroleum hydrocarbon (PHC) was determined in sediments from three different ecosystems such as Vellar and Coleroon estuary, Pichavaram mangrove and Parangipettai coast from the southeast coast of India. Regarding the composition of sediment texture, maximum value of sand 84.4% was noticed at Vellar estuary (V1) during post monsoon. The higher values of silt (51.5%) and clay (34.1%) were observed during pre monsoon and summer respectively in Parangipettai coast (PC1) at the exit point of Vellar estuary. The maximum value of TOC (6.2mg g^{-1}) was observed during post monsoon season at Pichavaram mangrove (PM1). The minimum value of TOC (1.0mg g^{-1}) was observed at in Vellar estuary (V1) during summer. The values of PHC in three ecosystems, and for all the seasons, the higher value of 5.6µg g^{-1} was observed during monsoon season in Parangipettai coast (PM3), at the exit point of Coleroon estuary. The lower value of PHC (2. 2µg g^{-1}) was observed during summer at Coleroon estuary (C3), the fresh water mixing zone. The PHC concentration from the different ecosystems was found in the following order Paragipettai coast > Pichavaram mangrove > Vellar & Coleroon estuary. The contamination factor (C_f) was applied to assess the ecological risk of PHC and was found to be < 1, indicating the study area is still pristine. It is also confirmed that the PHC values obtained from the present study were lesser than the natural background value of Persian Gulf sediments (10-15 µg/g) and therefore the study concludes that the Vellar and Coleroon estuary, Pichavaram mangrove and Parangipettai coast had low level of pollution in terms of PHC contamination.

Key words: Petroleum Hydrocarbons, Ecosystems, Estuary; Mangrove and Coastal contamination factor.

1. INTRODUCTION

The petroleum hydrocarbons (PHC) enter into the estuary, mangrove and coastal area is mainly from river runoff to the study area which carries the industrial discharges; automobile wastes, combustion of fossil fuels etc. PHC is hydrophobic, lipophilic and the accumulation in sediments is a potential risk for the organisms in future so, it is also considered as "Chemical Time Bomb". Studies pertaining to the Petroleum hydrocarbons (PHC) become inevitable because of their carcinogenic and mutagenic properties and affect the biological system through bioaccumulation and biomagnification. The world wide usage of PHC is tremendously increased [1] due to the vast development of the industries machineries, automobiles, shipping activities etc. Even though the natural process contribute to some extent of oil pollution to the coastal and near shore ecosystems yet anthropogenic source become more important because of the indiscriminate use of petroleum, petroleum by products and petrochemicals etc. Because of the growing influence of fossil fuel production and uses, hydrocarbon derived contamination has become of special concern. However, for abating inputs of hydrocarbons such as PHC which pose risk to animal and human health, it is essential to identify the major sources to take necessary control measures [2]. Petroleum hydrocarbon has received special attention because it is readily adsorbed onto particulate matter and bottom sediments ultimately act as a reservoir for hydrophobic contaminations. Sediment integrates the pollutants over time and it is easy for the

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sedimentary organisms to concentrate the pollutants in an alarming rate. Investigation on the composition of hydrocarbons in different marine sediments can provide much information about their source and diagnostic process and reflect the extent of anthropogenic pressure on the environment [3, 4, 5]. Immediately after the release of crude oil into marine waters, due to the hydrophobic nature, the petroleum compound (PHC) readily adsorbs on to the suspended particulate matter and accumulates in bottom sediments, and remains unchanged for long term; and pose serious threat to the structure of the benthic community [6, 7]. The concentration of PHC in the study area is mainly through the rivers such as Vellar and Coleroon and they are the important rivers in south east coast of India. For the present study, three different kinds of ecosystems were selected such as estuary, mangrove and coastal in order to obtain the base-line date on PHC and to understand the level of contamination.

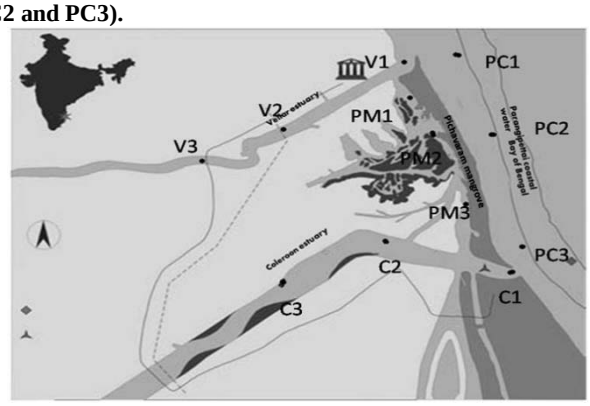
2. MATERIALS AND METHODS

Study area:

Vellar and Coleroon estuarine ecosystem:

The study has been conducted in Vellar (Lat. 11° 48' N; Long79° 76' E) and Coleroon estuaries (Lat. 11°25 'N; *L*ong 79°48' E) selecting three salinity gradient zones in addition to Pichavaram mangrove realm and Parangipettai coastal region. In estuaries, the salinity gradient zone is based upon the proportionate mix of fresh (river) and saline (marine) waters. In the mouth of the estuary, the salinity will be more due to the invasion of the sea water into the estuary whereas the salinity will be negligible at the fresh water mixing zone due to domination of fresh water influx. At the middle of the portion of the estuary the salinity will be moderate so the eco tone areas between marine and fresh water habitats in estuaries are characterized by the unique combination of physical, chemical and biological features, and are distinguished by exceptionally high productivity, Estuaries serve as both pollution sources for the coastal sea and marginal filter to the polluted runoff from the river drainage basin [8, 9] keeping the view in mind, 3 stations have been selected in each estuary according to various salinity gradient in which sea water mixing zone fresh water mixing zone and the zone of moderate salinity are included as shown in (**Fig 1**).

Fig 1: Map showing the sampling sites of Vellar estuary (V1, V2 and V3), Coleroon estuary (C1, C2 and C3), Pichavaram mangrove (PM1, PM2 and PM3) and Paragipettai coast (PC1, PC2 and PC3).



Pichavaram mangrove ecosystem:

The Pichavaram mangrove ecosystem (Lat 11° 26' N; Long 79° 77' E) in Tamil Nadu coast is very important in terms of ecological point of view and it is formed out of the network of channels and adjoining creeks through the estuaries of Vellar and Coleroon. This ecosystem fringed with variety of mangrove plants, mud flat shrubs which serve as significant breeding ground for number of marine and brackish water organisms. Three stations have been selected starting from northern end (mixing point of Vellar estuary) to southern end (mixing point of Coleroon estuary) as shown in (Fig 1).

Parangipettai coastal region:

The Vellar and Coleroon estuaries have opening into the Bay of Bengal and it is named after the Parangipettai Coastal region (Lat 11° 49' N; Long 79° 48' E). The sampling have been made at about 30mts depth between the exit point of Vellar and Coleroon estuaries, 3 sampling sites were selected covering even distance as shown in (Fig 1).

Sampling and analysis:

Collection of sediment samples

The sediment samples were collected from year of 2011 to 2012 the sampling sites covering all the four seasons summer (April), Pre monsoon (July), monsoon (October) and Post monsoon (January) using Peterson grab with an area of 0.2 m^2.The collected sediments sample preserved in pre - cleaned polythene bags with ice and brought to the laboratory for further analysis.

Sediment composition:

About 100 gm of dried sediment was sieved by mechanical shaker [10] to investigate the sediment composition.

The sand fraction was determined as the amount of sediment retained by sieve of 125mm size.

The portion which passes through the sieve of 125 mm but retained by sieve of 0.063mm is slit and the fraction which passes through the sieve of 0.063mm is clay.

Total Organic Carbon (TOC):

The surface sediment was separated (upper 2cm) from the grab for the analysis of Total Organic Carbon content. Total Organic Carbon content (TOC) was estimated using chromic acid oxidation method followed by titration with ammonium ferrous sulphate (Walkley – Black method) as modified by Gaudette *et al* [11].

Analysis of PHC in sediments samples:

The sediment sample was saponfined using KOH-methyl alcohol mixture. About 10 gram of sample was subjected for soxhlet extraction with n-hexane. Then the extracts were added with a required amount of anhydrous sodium sulphate and shaken well for the removal of moisture. The extract was concentrated down using rotary evaporation at 30°C followed by concentration with a nitrogen gas stream down to a volume of 1 ml. The PHC was measured using a UV-Spectrofluorometer (Cary Eclipse fluorescence spectrophotometer) at 360 nm emission after excitation at 310 nm. All blank, standards and samples were measured in a Teflon capped 1 cm silica fluorescence cell under identical instrumental setting and conditions. Duplicate spikes and blanks were treated identically Chrysene as a standard reference to test precision accuracy and solvent purity in the analytical procedure and the data were expressed in terms of chrysene equivalents. Percentage of recovery for spiked samples ranged from 94% to 98%, whereas precision agreed within 6%. Blank values were almost negligible. All experiments were conducted in 3 replicates and the average of the values was reported (IOC – UNESCO 1984).

Statistical evaluation:

Regression used for analysis of positive and negative correlation between the variables. One way ANOVA used for identify the significant level of temporal and spatial variations of variables using SPSS 16 [12].

3. RESULTS AND DISCUSSION

The composition of sediments:

The values of sand, silt and clay in the sediment samples of the Vellar and Coleroon estuaries, Pichavaram mangrove and Paragipettai coast are given in (**Table 1**). The ANONA for soil texture, TOC and PHC is given in Table 3. The significance level for clay, TOC and PHC was found for all the station throughout the study. The sand and silt were showing insignificant level. Among these three ecosystems, the values sand and silt were found with slight fluctuation when look into seasonally. The reason may be due to the continuous water flow between these ecosystems through tidal action. So the sedimentation process might be same in each ecosystem. The observed fluctuation among seasons might be due to the heavy flooding in the rivers during the monsoon season which contribute the influx of sediment considerably. Therefore the maximum value of sand (84.4%) was noticed at vellar estuary (V1) during post monsoon season. The minimum of 40.6% was measured during summer in the paragipettai coastal region (PC1). The higher values of silt (51.5%) and clay (34.1%) were observed during Pre monsoon and summer respectively at the exit point of vellar estuary in parangipettai coast (PC1). This again confirms that during monsoon period the rate of sand influx is slightly higher. The report on sediment composition made by Lyla *et al*. [13] and Joydas *et al*. [14] in the continental shelf region of south east of India and west coast of India respectively had agreed with the present study as the sediment composition is dominated with higher percentage of sand followed by silt and clay. The textural and depositional features can be assessed through sedimentalogical data. The order of sediment characteristic feature is as follows sand > silt > clay. The sand and silt occupies 10 to 80 % of total samples and the clay is observed between 1.5 to 34 %. These findings are having good agreement with the study made by Balakrishna Prasad *et al*. [15] in the same study area.

Total Organic Carbons (TOC) and Petroleum Hydrocarbons (PHC):

The values of Total Organic Carbon and Petroleum hydrocarbons are presented in Table 2. The variation of TOC and PHC of all the stations was observed at the 0.001 level of significance in Table 4. Considering the values of TOC in these three kinds of ecosystems, Pichavaram mangrove ecosystem is noticed with higher average values than other ecosystem. This may be due to the deposition of the enormous litter from mangrove plants. The maximum value of TOC (6.2 mg g^{-1}) was observed during Post monsoon season at station PM1 of Pichavaram mangrove. The minimum value of TOC (1.0 mg g^{-1}) was found

during summer at station V1 in Vellar estuary. The increased value of TOC at Post monsoon can be attributed to the sedimentation process which was taken place at monsoon season in addition to the continuous decay of litter from mangrove plants. The TOC values obtained in the present study for the same ecosystem shows much lower than the values obtained by the Balakrishna Prasad *et al*. [16]. This shows that the gradual deposition of organic matter on to sediment becoming lesser year by year, and it may be due to more deposition sand rather than the clay from the riverine system. In contrast to the present study, the study made by Hong-Gung Ni *et al*. [17] in coastal region off south china shows that the fluivial organic load from the reverine is more especially during flood events.

In the present study, temporal variations of TOC and PHC values were not found with significant regression as shown in Fig. 2. This indicates that the input of PHC to the study area was irregular and not in accordance with TOC levels. While analysing the values of PHC in all the ecosystems and for the seasons, the higher value (5.6 µg g^{-1}) was noticed during monsoon season at station PC3 of Paragipettai coast (exit point of Coleroon estuary) whereas the lowest value of 2.2 µg g^{-1} was measured during summer at the fresh water mixing zone (PC3) of Coleroon estuary. This type of distribution clearly indicates that the North east rainy seasons which influence the PHC concentration to such elevated levels by bringing the terrestrial waste through flood. The values of PHC in the sediments in the present study show good similarity to the values obtained by the Subramanian Veerasingam *et al*. [18] from the Tamil Nadu coast. They obtained higher values (4.23) at Narimanam and Nagapatinnam coast and lower value (1.48) at Parangipettai coast and the reason attributed to such values due to high proportion of sand in Parangipettai coast. The present study supports their study by showing higher amount of sand in the sediment proportions.

The average concentration of PHC measure by Ye *et al*. [19] in sediment sample from rivers and canals in Tianyin – China shows very high levels (0.072 to 3.000 mg g^{-1}) when compared with the present study. The study made by Zheng Richardson [20] also showed the exceeded levels of PHC (0.0045 – 1.996 mg g^{-1}) when compared to the present study. They explained for such enhanced level was due to many local inputs through the rivers and canals that lies in the upstream to the urban area. The average value of PHC in each ecosystem varies with smaller fluctuations. The following order was found out for each ecosystem with regard to contamination pattern as Estuary > Pichavaram mangrove > Parangipettai coast.

Contamination factor of Petroleum Hydrocarbons:

The concentration of PHC and its contamination factor in the sediments covering four seasons are presented in Table 4. In earlier report of PHC values in Pichavaram mangrove and Paragipettai coastal region ranged between 1.05 to 7.71 ppm [21] and their values are found to be similar to the present study. Venkatachalapathy *et al*. [22] reported higher values of PHC concentration in sediments from Chennai coast, India (1.88 – 39.76 ppm) when compared to the present study which shows that the modernisation and fast development of the metropolitan city contributes such higher levels. The reports made by Chouksey *et al*. [23] for PHC concentration in sediments from Ulhas estuary and Thane creek were ranged from 2.0 to 40.8 ppm and 7.6 to 42.8 ppm respectively which was comparatively higher to the present study. The report of PHC values from Persian Gulf sediments was categorized into four levels (guideline µg /g) i.e., Unpolluted area /natural background (10-15), slightly polluted / upper permissible limits (15-50), moderately polluted (50-200) and heavily polluted area (> 200). The contamination factor ($C_f = C_0\text{-}1/ C_n$) of PHC was used by Hakanson method, and the level of pollution are arranged as follows:

$C_f < 1$ low pollution, $1 \leq C_f < 3$ middle pollution, $3 \leq C_f < 6$ notable pollution, $C_f > 6$ high pollution- (C_0-1: present contamination, C_n: Natural background [24]. In the present study, in all the stations, the concentrations of PHC were less than the natural background value (10-15 µg/g) as represented by Massoud [25].

Table 1: Percentage (%) of sediment texture in pre monsoon (PM), monsoon (M), Post monsoon (PoM) and summer (S)

Station	Sand (%)				Silt (%)				Clay (%)			
	PM	M	PoM	S	PM	M	PoM	S	PM	M	PoM	S
V1	65.1	74.5	84.4	67.1	33.1	21.2	13.3	31.4	1.8	4.3	2.3	1.5
V2	57.0	57.9	62.2	54	31.1	32.7	22.5	34.1	11.9	9.4	15.3	11.9
V3	71.3	62.2	70.2	70.3	19.1	26.5	15.6	19.1	9.6	11.3	14.2	10.6
C1	69.6	69.7	75.6	70.1	27.3	26.4	19.9	27.6	3.1	3.9	4.5	2.3

C2	55.1	65.6	68.2	51.1	34.3	30.1	27.8	37.3	10.6	4.3	4	11.6
C3	52.5	64.3	62.4	51	38.3	26.5	23.3	37	9.2	9.2	14.3	12
PM1	61.1	65.1	59.5	60.1	16.0	22.6	16.3	17.3	22.9	12.3	24.2	22.6
PM2	49.1	48.5	52.8	44.1	25.8	25.1	20.4	26.4	25.1	26.4	26.8	29.5
PM3	47.6	49.2	45.7	46.3	22.3	24.1	18.3	21.3	30.1	26.7	36	32.4
PC1	45.4	67.7	42.5	40.6	51.5	26	50.5	25.3	3.1	6.3	7	34.1
PC2	79.2	69.4	70.7	73.2	11.5	26.4	23.4	24.6	9.3	4.2	5.9	2.2
PC3	65.3	61.5	55.5	64.5	27.6	35.3	39.3	30.9	7.1	3.2	5.2	4.6
Min	45.40	48.5	42.5	40.6	11.5	21.2	13.3	17.3	1.8	3.2	2.3	1.5
Max	79.20	74.5	84.4	73.2	51.5	35.3	50.5	37.3	30.1	26.7	36.0	34.1
Mean	59.86	62.97	62.5	57.7	28.2	26.9	24.3	27.7	11.98	10.1	13.2	14.6
SD	10.52	7.9	12.2	11.3	10.8	4.02	10.9	6.6	9.2	8.3	10.8	12.06

Table 2: Concentration of TOC and PHC in pre monsoon (PM), monsoon (M), Post monsoon (PoM) and summer (S)

Station	Total Organic Carbon (TOC mg/g)				Petroleum Hydrocarbons (PHC µg/g)			
	PM	M	PoM	S	PM	M	PoM	S
V1	2.7	2.1	2.1	1.0	3.1	4.1	3.7	3.0
V2	3.1	3.0	3.8	2.1	2.7	3.7	3.6	2.5
V3	2.6	5.1	4.2	2.1	3.0	3.9	3.5	2.6
C1	2.1	2.2	3.6	1.9	3.2	4.6	3.4	2.9
C2	3.4	3.2	4.5	3.3	2.5	4.2	3.1	2.6
C3	1.7	3.4	2.4	1.6	3.1	3.9	2.8	2.2
PM1	5.9	4.6	6.2	4.6	3.8	5.1	4.1	3.3
PM2	4.6	3.6	5.3	4.2	3.4	4.3	3.8	2.7
PM3	3.1	4.5	3.1	3.0	3.5	4.1	3.5	2.4
PC1	2.6	3.4	3.4	1.2	4.4	5.2	4.2	2.9
PC2	2.3	2.7	3.4	2.1	3.8	4.2	4.5	3.6
PC3	4.6	3.1	4.6	3.2	4.2	5.6	4.8	3.9
Min	**1.7**	**2.1**	**2.1**	**1.0**	**2.5**	**3.7**	**2.8**	**2.2**
Max	**5.9**	**5.1**	**6.2**	**4.6**	**4.4**	**5.6**	**4.8**	**3.9**
Mean	**3.2**	**3.4**	**3.9**	**2.5**	**3.4**	**4.4**	**3.8**	**2.9**
SD	**1.2**	**0.9**	**1.2**	**1.1**	**0.56**	**0.59**	**0.57**	**0.5**

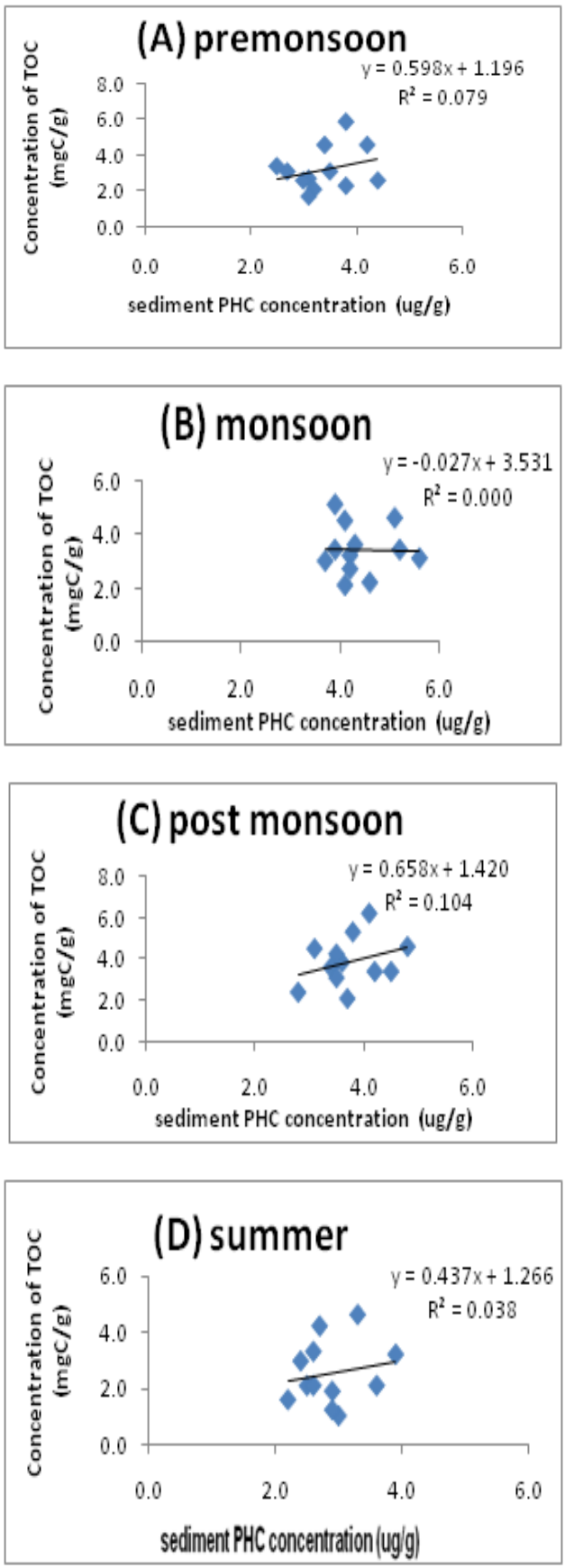
Table 3: ANOVA for soil texture, TOC and PHC

		Sum of Squares	df	Mean Square	F	Sig.
Sand	Between Groups	1268.712	3	422.904	4.777	.006
	Within Groups	3895.568	44	88.536		
	Total	5164.280	47			
silt	Between Groups	711.201	3	237.067	4.029	.013
	Within Groups	2589.077	44	58.843		
	Total	3300.278	47			
clay	Between Groups	3032.802	3	1010.934	27.057	.000
	Within Groups	1643.986	44	37.363		
	Total	4676.788	47			
TOC	Between Groups	17.632	3	5.877	6.144	.001
	Within Groups	42.091	44	.957		
	Total	59.723	47			
PHC	Between Groups	7.892	3	2.631	6.554	.001
	Within Groups	17.662	44	.401		
	Total	25.555	47			

Table 4: Mean concentration and contamination factor for PHC (µg/g)

Station	season	Mean PHC (µg/g)	Guideline*	Contamination Factor (C_f)
Vellar estuary	Pre monsoon	2.9	10-15	0.29
	monsoon	3.9	10-15	0.39
	Post monsoon	3.6	10-15	0.36
	summer	2.7	10-15	0.27
Total average		3.28	10-15	0.328
Coleroon estuary	Pre monsoon	2.9	10-15	0.29
	monsoon	4.2	10-15	0.42
	Post monsoon	3.1	10-15	0.31
	summer	2.6	10-15	0.26
Total average		3.20	10-15	0.320
Pichavaram mangrove	Pre monsoon	3.6	10-15	0.36
	monsoon	4.5	10-15	0.45
	Post monsoon	3.8	10-15	0.38
	summer	2.8	10-15	0.28
Total average		3.68	10-15	0.368
Paragipettai coast	Pre monsoon	4.1	10-15	0.41
	monsoon	5.0	10-15	0.50
	Post monsoon	4.5	10-15	0.45
	summer	3.5	10-15	0.35
Total average		4.28	10-15	0.428

Fig 2: Regression for TOC Vs PHC A) Pre monsoon B) monsoon C) Post monsoon and D) summer



CONCLUSION

The concentration of PHC in sediments collected from different interconnected ecosystem such as Vellar and Coleroon estuaries, Pichavaram mangrove and Parangipettai coast are very much lower when compared to natural background level can be considered as a low polluted region. The result of PHC obtained in the present study can be used for the future base-line studies. Even though the sediments showed the contamination factors and risk index of all the stations indicating below 1 (Cfv < 1) shows that the study area is not polluted, yet continuous monitoring is necessary because the study area is vulnerable and also situated in a strategic location that the major fishing activities and main rivers have downstream and have direct opening to the coast and mangrove ecosystem.

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