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Variability of Petroleum Hydrocarbons in the Industrial Effluent Discharge Influenced Coastal Region on the Central East Coast of India

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ABSTRACT
Total petroleum hydrocarbons (TPHC) and their seasonal variability are investigated in the coastal region influenced by industrial effluent discharge on the central east coast of India to study TPHC pollution in the coastal oceans. Our results suggest that the study region is not significantly polluted by TPHC during either pre-monsoon or post-monsoon seasons even with the release of treated effluent of ~305 MLD per week from 10 pharmaceutical and 1 textile industries. It could be due to relatively less TPHC input from industrial effluents and/or a better dilution of industrial effluents by the prevailing coastal currents and circulation. This study provides the first ever baseline dataset on TPHC in the coastal region impacted by industrial effluent release. These results can be used to understand the impact of industrial effluent in the coastal regions, and for formulating the strategies for release of industrial effluents into sea by the policy makers.

Key words
Petroleum hydrocarbons, central east coast of India, industrial effluent, seawater quality, pollution.

1. INTRODUCTION
Petroleum hydrocarbons (PHC), a group of hundreds of chemical compounds and potential pollutants [41], in coastal regions mainly derive from anthropogenic activities such as incomplete combustion of petroleum products and coal, biomass burning, river run off, atmospheric deposition, release of urban sewage and industrial effluents, oil production, refining and its transport through marine traffic, and shipping and boating activities [6, 8, 10, 48] besides biogenic origin [42]. In the marine environment, it was estimated that about 10% of the crude oil comes from natural seeps, 27% from production, refining and transportation of oil and 63% from river runoff, domestic and industrial sewage and atmospheric deposition etc. [29]. Coastal habitats are relatively more exposed than open ocean habitats to petroleum hydrocarbons [19, 21] because of higher concentrations of total petroleum hydrocarbons (TPHC) by about 1000 times in the coastal region compared to those in the open sea due to the transport of contaminants from land is more to the coastal than to the open ocean regions [7]. Major sources and their contributions to TPHC in coastal oceans are widely variable in space because of differences in the proximity of location to the regional PHC sources and the magnitude of contamination from different sources [9, 47]. Higher levels of PHC in aquatic systems cause bioaccumulation in soft tissues of marine organisms (zooplankton), fish, mussels, corals and mollusks etc [3, 5, 22]. Since many of the PHC compounds are carcinogenic and mutagenic in nature [35], and their bioaccumulation severely affect the aquatic ecosystem and subsequently the human health. Studies on PHC in seawater and aquatic organisms are thus of global importance.

Although, petroleum hydrocarbons were recognized as pollutants in the early 1950s, our understanding on their concentrations and major sources in estuaries and coastal regions of the world is poor due to the paucity of data from many regions. Though most of the studies conducted were confined to North America and Europe [16, 30, 39, 57], studies on coastal regions of Southeast Asia are scarce. Further, levels of PHC in the industrial effluent dominated coastal regions are unknown.

Very limited studies were conducted on petroleum hydrocarbons in the Indian coastal waters but most of them were confined to the west coast of India, eastern Arabian Sea [8, 12, 24, 45]. A few studies were conducted in the sediments on the east coast of India, the western Bay of Bengal [33, 53, 54]. Dissolved/dispersed TPHC concentrations in the coastal waters of east coast of India and the impact of treated industrial effluent released into the sea through Marine Outfall Points (MOPs) on the TPHC levels in this region are unknown.

The main objective of this study is to investigate the effect of industrial effluents released on TPHC in the coastal region of north Andhra coast, central east coast of India. This is one of the best coastal locations to examine the impact of industrial effluent on TPHC in coastal regions because 11 major industries discharge a total effluent of ~305 MLD per week into this 150 km long stretch of the coastal region.

2. MATERIALS AND METHODS
A total of 148 sampling stations were occupied at and around the MOPs of various Industries (see Figure. 1) and off major ports for in-situ observations and sample collection on board a big mechanized fishing trawler. Field campaigns were conducted during both pre-southwest monsoon (17 February to 08 March 2018) and post-southwest monsoon (6–31 December, 2018) seasons to understand the seasonal variability of TPHC in the study region. Samples were collected during the day prior to effluent discharge and during the day of discharge at the MOPs

of industries, and at stations 0.5, 1.0 and 2.0 km away in all four directions (0^o, 90^o, 180^o and 270^o) from the MOP of each industry in order to assess the impact of effluent release on TPHC in the study region. Samples were collected from surface and near bottom for dissolved oxygen (DO), phytoplankton biomass (chlorophyll-a), dissolved inorganic nutrients (ammonium, nitrite, nitrate, phosphate and silicate), TPHC and total suspended matter. Samples for TPHC were collected in pre-cleaned amber coloured glass bottles (1L) and acidified (pH <2) using 6 N hydrochloric acid and kept at 4^oC until transferred to the shore laboratory on the same day.

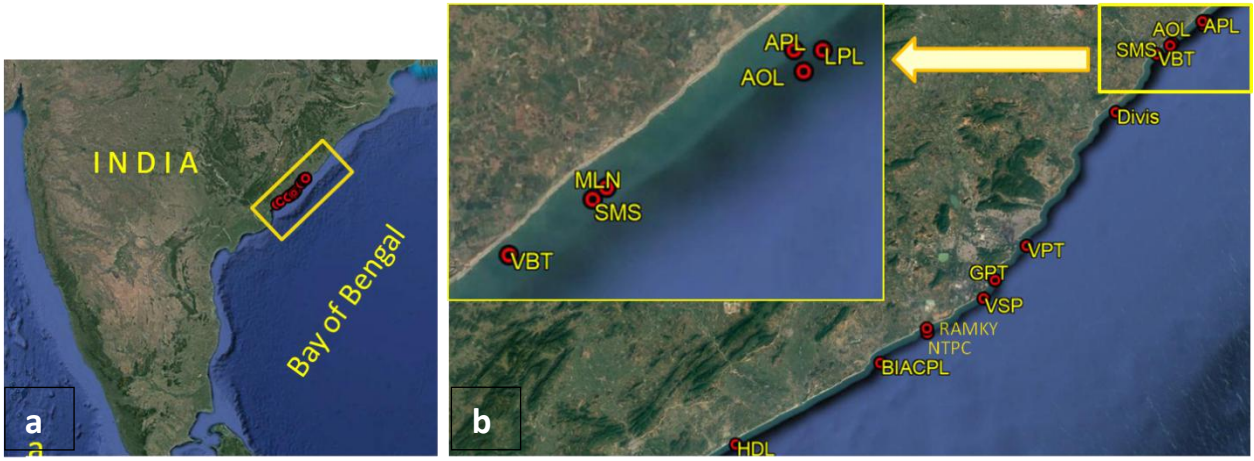

Figure 1. (a) Map showing the study region in the central east coast of India. (b) Map showing marine outfall points (MOPs) of Industries through which effluent discharges into the study region. Region shown by rectangle was enlarged to show all MOPs in the region and provided in the inset picture. APL: Aurobindo Pharma Ltd., AOL: Andhra Organics Ltd., LPL: Lantech Pharmaceuticals Ltd., MLN: Mylan Laboratories Ltd.; SMS: SMS Pharmaceuticals Ltd., VBT: Vizianagar Biotech Ltd., Divis: Divis Laboratories Ltd. VPT: Visakhapatnam Port, GPT: Gangavaram Port, VSP: Visakhapatnam Steel Plant; RAMKY: JN Pharma City Ltd., NTPC: National Thermal Power Corporation, BIACPL: Brandix India Apparel City Pvt. Ltd., HDL: Hetero Drugs Ltd., DFC: Deccan Fine Chemicals Ltd.

Water column temperature and salinity were obtained from conductivity-temperature-density (CTD) profiling system (Sea Bird Electronics, SBE19 plus). Dissolved oxygen was determined by Winkler's titration method using potentiometric end point detection system (Metrohm, Switzerland). Dissolved inorganic nutrients were analyzed by spectrophotometric method using autoanalyzer (Skalar, The Netherlands). Repeated analysis of aliquots of samples and standards results in the precision, in terms of standard deviation, of 0.07%, 0.02µM, 0.02µM, 0.01µM and 0.02µM respectively for DO, ammonium, nitrate+nitrite, phosphate and silicate respectively.

TPHC concentrations in seawater samples were determined by a standard liquid-liquid extraction method (LLE, EPA method 3510) [4, 34, 36, 51] using Ultra Violet Spectrofluorometric (UVF) detection technique [18, 50, 56] which is more efficient and reliable for TPHC determination in water samples. Seawater sample (500ml) was extracted with HPLC grade n-hexane (20ml) three times and the combined extract was dried over anhydrous sodium sulphate to remove moisture content. Fluorescence of the extract was measured at an emission wave length of 360 nm (excitation wave length 310 nm) using spectrofluorometer (Cary Eclipse, Varian). Blanks prepared by following the same procedure which was employed for sample collection were used to correct the fluorescence of the samples. PHC concentrations in seawater were calculated from the multi-point calibration established by chrysene as a standard. Results of TPHC concentrations in seawater samples are expressed as chrysene equivalents. Repeated analysis of aliquots of samples and standards yielded ±4% of the precision for the TPHC measurements.

3. RESULTS AND DISCUSSION

Phytoplankton biomass, in terms of chlorophyll-a, in surface waters varied from 0.1 to 7.5 mg m-3 (mean 2.4±1.7 mg m-3) and from 0.2 to 4.1 mg m-3 (1.1±1.0mg m-3) during pre- and post- southwest monsoon seasons respectively. Corresponding DO concentrations in the surface waters ranged from 4.4 to 7.8 mg l-1 (mean 6.0±0.5 mg l-1) and from 5.9 to 8.5 mg l-1 (7.3 ±0.5 mg l-1) respectively. Similar trend was observed in the bottom waters, with mean higher concentrations of Chl-a and lower concentration of DO during the pre-southwest monsoon (2.8±1.9 mg m-3 and 5.4±0.7 mg l-1, respectively) than during the post-southwest monsoon season (1.0±0.7 mg m-3 and 7.1±0.4 mg l-1, respectively). Relatively higher Chl-a is noticed during the pre-southwest monsoon than that of the post-southwest monsoon due to the occurrence of coastal upwelling (close to coast) at the central east coast of India during the pre-southwest monsoon season (February-March). Coastal upwelling occurs due to weaker salinity stratification by strong alongshore north-eastward winds and poleward flowing east India Coastal Current [37, 44, 49] that bring nutrient-rich sub-surface waters to surface and enhances primary production during this period. Elevated levels of primary production during April-May were also reported along the western coastal Bay of Bengal [15, 40] due to strong coastal upwelling in association with a well-developed poleward western boundary current [13, 37, 43]. Concentrations of dissolved inorganic nutrients, Chl-a

and DO in the study region are consistent with those found in the coastal regions of east coast of India which are not influenced by industrial effluent discharge, for example, off Kakinada. This suggests that no proliferation of phytoplankton biomass takes place due to the supply of nutrients through the release of industrial effluent in the study region during either pre- or post-southwest monsoon seasons.

TPHC concentrations in surface waters varied from 6.5 to 56.1 µg l-1 and from 0.5 to 165 µg l-1, with mean values of 21.3±11.4 µg l-1 and 20.6±26.7 µg l-1, during pre- and post-southwest monsoon seasons respectively. Corresponding TPHC concentrations in the bottom waters ranged from 6.5 to 52.6 µg l-1 (mean: 19.9±10.2µg l-1) and from 1.2 to 98.6µg l-1 (16.7±18.3 µg l-1) respectively. Mean TPHC concentrations found in this study are similar to those observed along the northwest coast of India [24]. Only three stations, i.e., near the mouth of the Gangavaram port (165 µg l-1), north of the MOP point of Brandix India Apparel City (132 µg l-1 and 99.5 µg l-1) and at the MOP point of Vizianagar Biotech Pharma Industry (124 µg l-1) recorded elevated levels during the post-southwest monsoon season. However, our TPHC concentrations are relatively lower than those found in many coastal regions of the world, for instance, Algoa Bay, South Africa (45 to 307 µg l-1). Comparison of TPHC in our study region with TPHC in coastal regions elsewhere in the world determined by the same method (UVF) is shown in Table 1.

Table 1: Comparison of TPHC concentration found in this study with the TPHC concentrations found in coastal regions elsewhere in the world determined by UVF method.

S. No.	Region	TPHC concentration (µg l^{-1})	Reference
1	Algoa Bay, Eastern Cape, South Africa	45 – 273	Abiodun O Adeniji et al., 2017 [2]
2	Coastal waters, Malaysia	320 – 2280	Abdullah, 1995 [1]
3	Gulf of Thailand	76	Wattayakorn et al., 1997 [55]
4	Bohai Bay, China	24 – 508	Li et al., 2010 [32]
5	North-western Arabian Gulf	1.2 – 546	EI Samra et al., 1986 [11]
6	Kara Sea	8 – 408	Korshenko et al., 2005 [26]
7	Central east coast of India	21.3±11.4 and 20.6±26.7	Present study

Statistically significant (two-tailed homoscedastic student t-test; $p<0.001$) seasonal variability was found in TPHC with relatively high concentrations during pre-southwest monsoon than during post- southwest monsoon. This could probably be due to dilution of TPHC by large amounts of freshwater runoff to the Bay of Bengal during the southwest monsoon (June-September). Since the Indian subcontinent receives >80% of its annual precipitation only during the southwest monsoon season, most of the annual freshwater discharge into the Bay of Bengal from the Indian rivers (6.023 x 1023 m3) occurs during this period [52]. Intense precipitation and freshwater runoff might have diluted TPHC concentrations during the southwest monsoon leading to lower concentrations during the post-southwest monsoon season.

In general, coastal waters containing TPHC concentrations of <1 µg l-1 are considered as unpolluted [6, 30] whereas TPHC concentrations exceeding >2 µg l-1 are considered as polluted [25]. However, Oppenheiner (1980) reported that TPHC concentrations of >100 µg l-1 should be considered as polluted. Harmful effect on aquatic organisms was found if seawater TPHC concentrations exceed 50 µg l-1 [17]. Nevertheless, the degree of TPHC contamination in coastal regions is highly region specific, and can vary from a few µg l-1 to few tens of mg l-1, based on proximity of the location to PHC sources [9, 47], such as harbours, ports, oil production sites, refineries, tanker traffic and industrial sewage etc. Because of this reason, the threshold limit of TPHC in coastal waters varies from country to country, for instance, Egyptian law of environment (50 µg l-1, [48]), the State of Alaska Department of Environmental Conservation (10 µg l-1 [4]), China and Russian Federation (50 µg l-1 [46]) and central pollution control board (CPCB) of India (100 µg l-1). European Union set a TPHC limit of 300 µg l-1 for estuaries and harbours.

Mean TPHC in seawater concentrations in the present study region during the pre-southwest monsoon (21.3±11.4µg l-1 and 19.9±10.2µg l-1 in the surface and bottom waters, respectively) and post-southwest monsoon seasons (20.6±26.7µg l-1 and 16.7±18.3µg l-1 in the surface and bottom waters, respectively) are far less than the threshold limit of Central Pollution Control Board (CPCB) and the limit of Environmental (Protection) Rules (1986) of India for oil and grease in the ecologically sensitive zones including shell fishing and salt pans (100 µg l-1) in the country. Though the present study region is influenced by the treated effluent release from 11 pharmaceutical industries, which discharges ~305 MLD of treated effluent per week, the relatively low TPHC in the study region could probably due to attaining sufficient dilution of effluent or sinking to sediments through mineral particles. The petroleum hydrocarbons have a tendency to adsorb on surfaces of the suspended mineral particles due to their hydrophobic properties [38]. Suspended particulates are, therefore, act as carrier of petroleum hydrocarbons from seawater to sediments [20]. In the present study, TPHC concentrations did not show significant relationship with TSM during either pre-southwest monsoon or post-southwest monsoon season, indicating that sinking of PHC to sediments through the suspended particles may not be the controlling mechanism of TPHC distribution in the study region. Phytoplankton biomass, in terms of Chl-a, was reported to decrease TPHC in surface waters of the Baltic Sea, particularly during the bloom periods, as phytoplankton absorb PHC and sinking to sediments [27, 28, 31]. However, TPHC concentrations, in the present study region, did not show significant correlation with phytoplankton biomass (Chl-a)

either during pre-southwest monsoon or during post-southwest monsoon, suggesting that the removal of PHC from surface waters by phytoplankton is not significant in the study region.

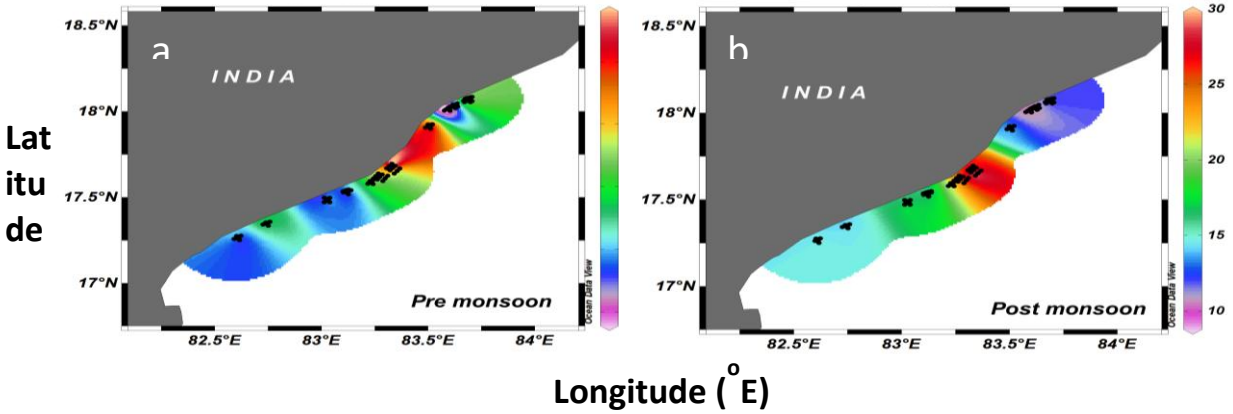

Figure 2. Distribution of total petroleum hydrocarbons (TPHC, µg l-1) in surface waters of the study region during (a) pre southwest monsoon and (b) post southwest monsoon seasons.

Attaining proper dilution of effluent in the coastal region after release also controls TPHC in the industrial effluent discharge dominated coastal regions as less dilution due to weak currents, tides or sluggish movement of water accumulates TPHC in the MOP regions. As per regulations of CPCB of India, it is mandatory that the treated effluent should attain at least 150 times dilution after its release at MOPs. More or less similar TPHC concentrations at MOPs in the coastal region during the day prior to effluent release (23.2±7.7µg l-1) and during the day of effluent release (mean: 20.5±8.4 µg l-1) (Table 2) indicates that the released effluent is diluted after its release. No significant (t-test; p=0.48) difference in concentrations of TPHC between the day prior to effluent release and during the day of effluent release (Table 2), suggest that the impact of industrial effluent release is not significant on TPHC contamination in the study region.

Table 2. Total petroleum hydrocarbon (TPHC) concentrations prior to the day and during the day of effluent discharge at some stations in the study region.

Station Name	PHC concentration (µg l^{-1})	
	Prior to the day of effluent discharge	During the day of effluent discharge
APL-MOP	26.3	25.0
AOL-MOP	25.8	9.1
SMS-MOP	22.6	26.0
MLN-MOP	9.0	20.2
VBT -MOP	28.4	6.5
Divis MOP	12.3	19.7
VSP-MOP	25.3	20.2
NTP-MOP	33.1	24.3
RMY-MOP	26.1	33.5
MOP: Marine outfall point. Station name codes are given in figure 1.		

4. CONCLUSION

Lower TPHC concentrations in the study region may also be due to significant influence of the other removal processes of petroleum hydrocarbons such as volatilization, microbial degradation and photo-oxidation and/or less PHC input from land etc. Further studies are required to evaluate the influence of various mechanisms controlling TPHC concentrations in the region. Our results indicate that the study region is not significantly polluted by TPHC. This study thus provides evidence that industrial treated effluent release into the sea through the MOPs, which are specifically designed based on the estimated effluent dilution factor in the sea, need not always cause TPHC pollution in the coastal regions.

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