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Urban Effluent Discharge into Rivers; A Forensic Chemistry Approach to Evaluate the Environmental Deterioration

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Abstract: Development of urban area provides deterioration of natural resources in the environment. Aliphatic hydrocarbons are among important chemical that show spatial changes. Fifteen surface sediment samples were collected using Ekman dredger to monitor the features of aliphatic hydrocarbons in tributaries of Likas River on December 2011. Samples were extracted using Soxhlet, followed 2 steps column chromatography then injected into GC-MS for instrumental analysis. The results show that northern tributary remained natural with odd carbon number dominance. The study has found fresh petroleum input into Inanam River where more urban development and transportation activities are existed by presence of major hydrocarbons such as C_{18} and C_{20}. The values of Carbon Preferences Index indicated that natural hydrocarbons entry from land is decreasing towards estuaries where marine input increases. Construction, transportations and urban activities around southern tributary of Inanam River have deteriorated drastically the quality of the environment. The study concluded that aquatic environments such as river are susceptive to anthropogenic activities. This research can scientifically monitor new residential developments environmental effects happening at the northern part of Darau River in the study area. The current approach may be employed to observe the rehabilitation programs in the environment.

Key words: Aliphatic Hydrocarbon · Surface Sediment · Major Hydrocarbons · Likas River · Deterioration

INTRODUCTION

Urban raw effluent contains vast verity of chemicals such as ions, metals, organic matters, oil and grease. The presence of these compounds in aquatic environment cause drastic drop of dissolved oxygen and production of CO_2, CH_4, H_2S and NH_3 result in acidity and aquatic organisms' mass mortality throughout aerobic and anaerobic bacterial processes. Among above chemicals, petroleum materials are dominant in any residential district due to often usage of fossil fuels in vehicles. Few developed countries such as Germany may collect and treat the daily wash-out from the city surface while the majority of countries fail to do that due to the cost of treatment operation and maintenances. Thus, these materials can always be found in various environmental compartments such as organisms [1].

Petroleum and oil products include vast verity of compounds such as aliphatic hydrocarbons that have negative impact to human and the environment. Aliphatic hydrocarbons (n-Alkane) are delivered from anthropogenic and natural sources. In the nature, the simplest form of alkane in which called methane forms by microorganisms [2, 3]. A wide variety of hydrocarbons including saturated alkanes contains 17 to 78 percent of heavy to light crude oil [4, 5]. Cracking is a process that breaks down heavier saturated hydrocarbons to shorter and consequently lighter hydrocarbons in which provides the source of energy in vehicles. Thus pollution from the urban application of petroleum products is tracked in their

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molecular fingerprints. Anthropogenic hydrocarbons including crude oil are well known as environmental pollutants [6]. United Nation Environment Program in 1992 has been released a guideline indicating a non-polluted area demonstrates the concentration of hydrocarbon by less than 10 ng/mg in sedimentary environment [7] however the observed concentrations often exceed this amount [8].

Normal alkane consist of odd and even carbon numbers ranging up to 70 carbons with no alkyl branch or substitutes [4]. The "Odd" carbon numbers are more abundant in biogenic sources anthropogenic sources provide either both of higher percentage of even carbon numbers. The hydrocarbon pollution was reported frequently from urbanized centers that travel by rain wash off force [9].

In this molecular marker approach several mathematical ratios are applied to identify the sources and origins of pollution such as Unresolved Complex Mixture (UCM), Carbon Preference Index (CPI), Major Hydrocarbon (MH), Average Carbon Chain (ACL) and C_{31}/C_{19} among others. The UCM appears in chromatograms from the Gas Chromatography machine that indicates the presence of old petroleum hydrocarbons [7]. The CPI is indicator for dominance of natural vs. anthropogenic hydrocarbons was introduced by [11] where widely used in later publications [12-15]. The high CPI values ranging from 3 to 6 demonstrate terrestrial vascular plants while oil pollution depletes the value up to unity [12, 13]. The ACL values fluctuate in presence of petroleum pollution where more polluted sites show depleted values [16]. Specific compounds from normal alkane provides clear indication of various sources such as terrestrial and vascular plants (C_{25}-C_{35}) [17], marine input (C_{19} and C_{17}) [18; 19] and oil pollution (C_{16}, C_{18} and C_{20}).

This study aims to investigate if urban untreated effluents affect characteristics and concentration of the aliphatic hydrocarbons. Two major rivers of Darau and Inanam in Likas district of Kota Kinabalu Sabah were chosen to represent a clean and polluted water bodies.

MATERIALS AND METHODS

Sampling: The study area is shown in Fig. 1. The Table 1 shows detail information of the studied site. The samples were aken using Ekman grab. Samples placed into pre-cleaned glass jars, shipped to the laboratory in cold condition and kept frozen in the freezer with lower than-20°C.

Cleaning of all equipment and laboratory glassware were done using tab and distilled water, analytical grade Methanol (MeOH) and Acetone and GC grade Hexane. Washed gears placed in a drying oven for 2 hours in 64°C prior to use.

Reagents and Standards: Normal alkane standards including n-C_{16}, C_{18}, C_{20}, C_{22}, C_{24}, C_{26}, C_{28}, C_{32} and C_{36} were purchased from Sigma Chemical Company, St. Louis, MO. A 100-200 (μm) mesh Silica Gel (F.C. 923) was used for column chromatography.

Fig. 1: Study area; circles in red show the sampling locations

Table 1: The features of the study area

Study Site	Stations	Latitude	Longitude	Depth (m)	Description
Darau River	1	6 00 47.60	116 06 45.10	0.60	Estuary
	2	6 00 47.80	116 06 51.00	0.30	Near Estuary
	3	06 01 07.80	116 07 04.60	3.10	Under KF bridge
	4	06 01 22.80	116 07 07.30	3.50	Near fish farming
	5	06 01 46.00	116 07 22.40	2.90	Residential development
	6	06 01 23.70	116 08 09.20	0.90	Upstream
Inanam River	1	6 00 47.60	116 06 45.10	0.60	Estuary
	2	6 00 47.80	116 06 51.00	0.30	Near Estuary
	3	06 00 35.60	116 06 54.5	0.80	Temporary settlement
	4	06 00 20.00	116 07 02.7	1.50	Temporary settlement
	5	06 00 12.12	116 07 08.4	1.50	Under the bridge of ring road
	6	05 59 26.50	116 07 12.5	0.30	Upstream

Experimental: The analytical techniques were described elsewhere [15]. In brief and with modification, sediment samples were freeze dried and an aliquot of 15 g of samples was transferred into the cellulose thimble and placed into the Soxhlet extraction. The Soxhlet was run for 11 h using Dichloromethane (DCM). The extract was purified and cleaned using first step column chromatography (9 cm and 0.9 cm i.d. using 20 mL of 3:1 Hexane: DCM). Then the sample was fractionated using second step column chromatography (18 cm and 0.47 cm i.d. using 4 mL of Hexane for n-Alkane). The extract that contained normal alkane was injected into the GC-MS machine.

Instrumentation: Normal alkanes analyzed using Perkin Elmer Clarus 500 Gas Chromatograph (GC) that coupled with Mass Spectrometry. A JandW Scientific Durabond DB-5, 30 m fused capillary column, 0.25 mm i.d. and 0.25 ìm film thicknesses. The helium gas used as mobile phase at 200 kPa. The injection port maintained at 300°C during the instrumentation. Oven and column temperature holds at 70°C for 1 min then planned at 30°C/min to 150°C, 5°C/min to 310°C and hold for 10 min. The detector temperature remains at 310°C.

RESULTS AND DISCUSSION

The results of the specific compound analysis of aliphatic hydrocarbon in this study are presented in Figure 2 (a and b).

The concentration of aliphatic hydrocarbon in specific compound basis shows that the river with untreated urban effluent demonstrates more than thousand folds of hydrocarbon comparing with clean river. Abundance of aliphatic hydrocarbon in lower molecular weight (C_{18} to C_{22}) and extreme higher molecular weight (C_{30} to C_{36}) were observed in upstream of polluted river as stations 5 and 6. LMW compounds indicate the freshness of the hydrocarbons while HMW usually derives from aged and/or burning processes.

The presence of odd and even carbon numbers is leading the natural and anthropogenic input of the hydrocarbon load into the environment [11]. Odd carbon numbers usually appear in lower concentrations than even numbers [20, 21] especially near the developed area (Figure 3).

As it is shown in Figure 4, the concentration of n-alkane fluctuates among stations, the highest concentrations of n-alkane were observed in stations 6 upstream of Inanam River followed by the estuary.

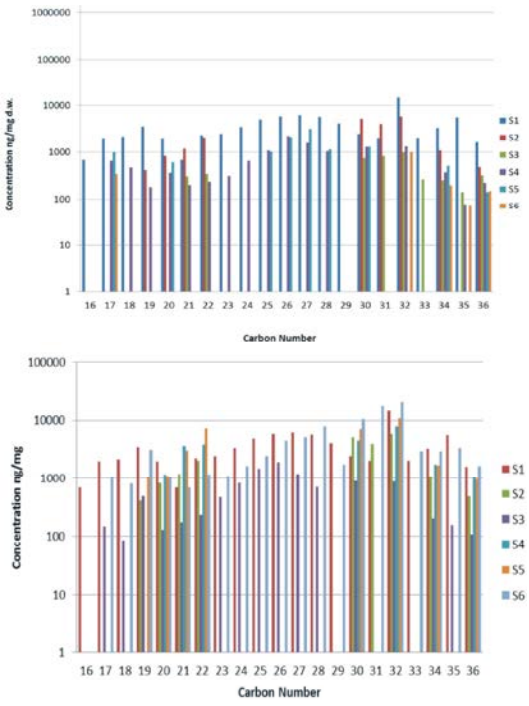

Fig. 2: Specific aliphatic hydrocarbon compound analysis in Darau River (up) and Inanam River (down).

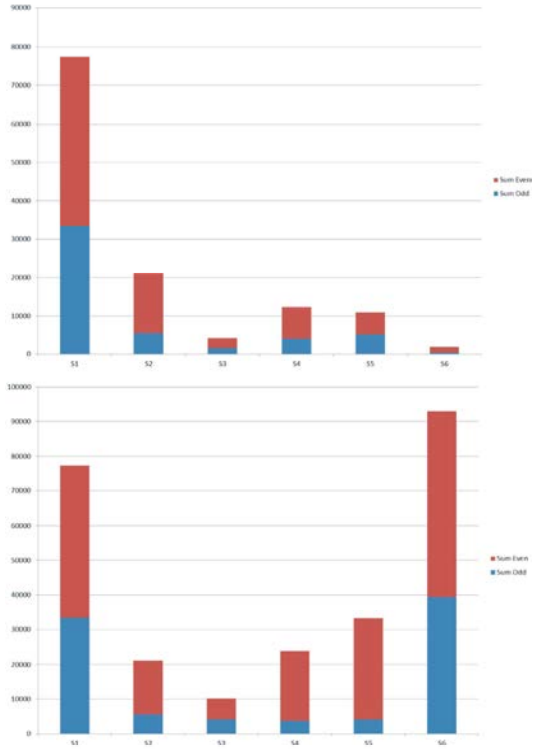

Fig. 3: Odd vs. Even carbon numbers in Darau River (up) and Inanam River (down). Station 1 is estuary and station 6 is upstream.

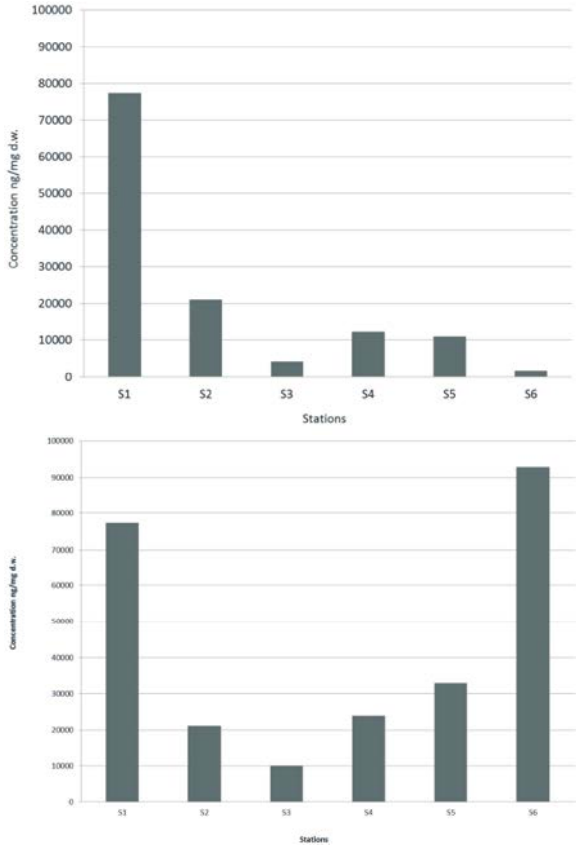

Fig. 4: Total n-alkane concentrations in Darau River (up) and Inanam River (down). Station 1 is estuary and station 6 is upstream.

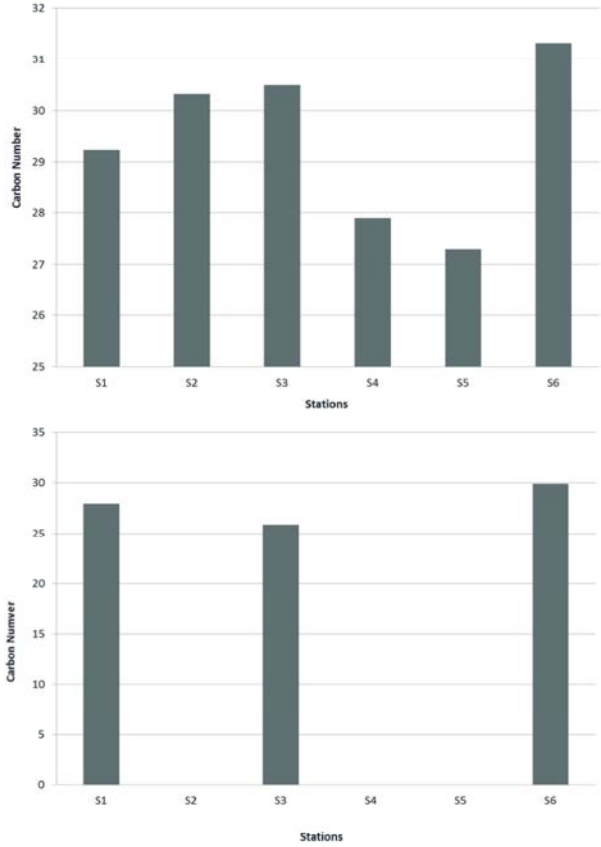

Fig. 5: The value of ACL in the study area; Darau River (up) and Inanam River (down).

The finding of this study is significantly higher than those reported by Hostettler *et al*. [22], Tolosa *et al*. [23], Commendatore and Esteve [24] and Gao *et al*. [25].

The results are compared with previous studies across Peninsular Malaysia where shown that Inanam River is 10 folds more polluted. The station S3 in the study area that demonstrate the least concentration of n-alkane has shown almost equal amount of pollution (c.a. 10,000 ng/mg) with the highest in Prai Straits near Penang Island in Peninsular Malaysia [26]. This is again higher than those concentrations of n-alkane of late 20th and early 21st century record in Johor Straits between Singapore and Peninsular Malaysia. The amount pollution in rivers usually are higher that marine environment since limited volume of water receive infinite amount of pollutants especially from the urban area. The same scenario was observed where middle stations in this study are compared with locations studied in the east coast of Peninsular Malaysia of Kelantan River, Terrengganue River and Besut River [26].

Average Carbon Chain Length Index (ACL): The application of ACL is a method to identify whether an environment is in a different chemical characteristic than others [16]. This tool were used before in several studies is identical to generally observe differences in among different ecosystems [27, 28]. The value is calculated using the following formula.

ACL value = [(25*C25)+(27*C27)+(29*C29)+(31*C31) +(33*C33)]/(C25+C27+C29+C31+C33)

As shown in the above formula, the values depend on the concentration of certain odd numbered hydrocarbons including C_{25}, C_{27}, C_{29}, C_{31} and C_{33}. These hydrocarbons are natural and consequently susceptive to environmental pollutants such as oil and industrial effluents [29]. The value of ACL in this study is shown below (Figure 5).

As shown in Figure 5, Inanam River demonstrates unstable ACL values where in station 2, 4 and 5 the ACL is mathematically omitted due to zero observation of certain odd carbon numbers. This performance is interpreted as

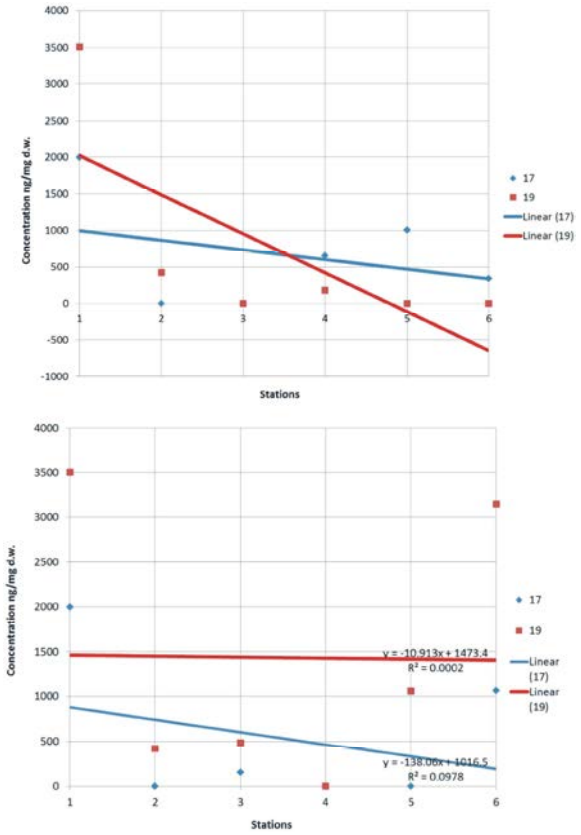

Fig. 6: Marin and aquatic input; Darau River (up) and Inanam River (down).

the effects of pollutants that results absence of odd carbon numbers [30, 31]. In comparison, values of ACL in Darau River are existed but fluctuating from approximate number of 27 to 31. The fluctuations among numbers in various stations are indicative for the minor effects of anthropogenic sources such as pollution, construction and agricultural activities. Darau River comparing to Inanam River has recently faced developments in housing and lands. A big residential area is being constructed in the near watershed where huge land clear cut and consequently wash out happens. Since city surface run-off and wastewater pollutants are not being discharged such as happening in Inanam River, this is believed to be observed a better ACL in Darau River.

Influence of Natural Hydrocarbons from Marine and Aquatic Sources: Certain hydrocarbons are indicative of natural activities from various sources. While higher odd numbered hydrocarbons such as C_{31} to C_{35} are indicative of terrestrial plants input, other including C_{17} and C_{19} represents marine microorganisms and aquatic plants.

The results of these hydrocarbons are shown in Figure 6 for both Darau and Inanam Rivers. In Figure 6 (a and b), the concentrations of C_{17} and C_{19} in presented in Y axis fluctuating up to 3500 ng/mg of dry weight in both rivers. Stations are in X axis plotted against the concentrations at Y axis.

In general, marine input decreases when moving from the estuary towards the upstream in both rivers which is considered a natural process throughout the monitoring of C_{17} hydrocarbon and depleting of marine chemistry such as salinity. The possible interpretations over the observation either the study area is highly polluted therefore less marine biogenic sources are available or it is subjected to rapid degradation after deposition as well as environmental limitation such as salinity [32, 33]. This is consistent with report from aliphatic hydrocarbons in Johor Straits in Peninsular Malaysia [26]. Aquatic plant input was monitored by monitoring of C_{19} hydrocarbon where Inanam and Darau Rivers show different patterns. In Darau River, it is depleting similarly to C_{17} while moving upstream however Inanam River shows a steady line while individual locations perform different. A high concentration of C_{19} was found exceptionally in Station 6 of Inanam River that supposed to follow the pattern from the other river. This observation is interpreted as deposition of huge amount of aquatic plant sourcing in upstream. This study was not able to conduct sampling in station upper than Station 6 since the river passes through residential area where municipality has developed an aqueduct to facilitate the urban irrigation. Meantime, the duct hosts infinite amount of algae and plant in which growing in presence of nutrients discharged from the urban area. Sudden flush following the heavy rain provides transfer of plants' materials into the first possible place of entrapment. Station 6 on Inanam River in this study has shown a trap of transported materials from upper stream while the same phenomena happens in the estuary as indicated in fingerprinting of C_{19} hydrocarbon.

Oil Sourced Anthropogenic Hydrocarbons: To monitor the oil sourced hydrocarbon, this research has employed certain compounds such as C_{16}, C_{18} and C_{20}. These 3 hydrocarbons are identical in recognition of fresh input of oil derivatives. The presence of this group is monitored by fingerprinting in gas chromatography-mass spectrometry machine. The concentration of each characterized named hydrocarbons are plotted against stations in Figure 7. The concentrations are presented in logarithmic distribution. Integration line is inserted for both locations with regression (R^2).

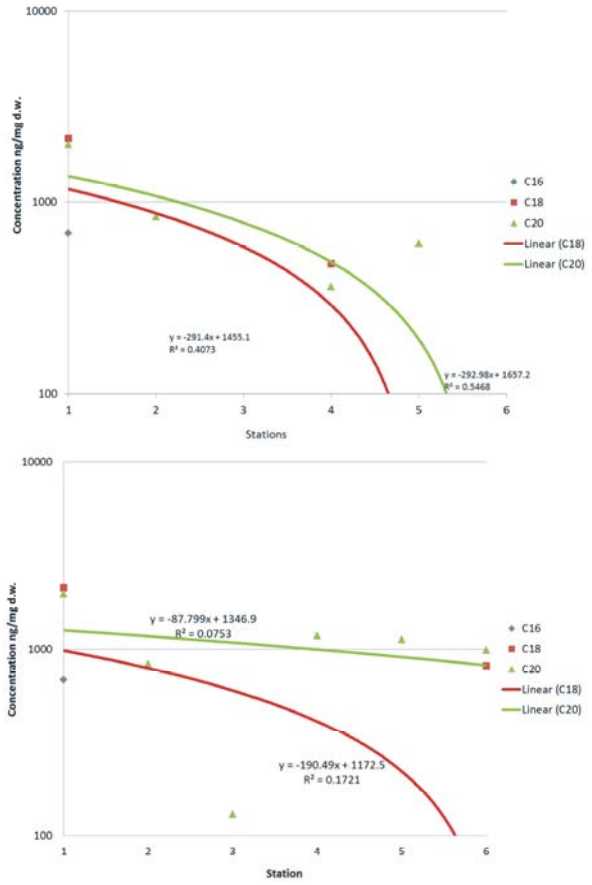

Fig. 7: Oil and oil derivatives input; Darau River (up) and Inanam River (down).

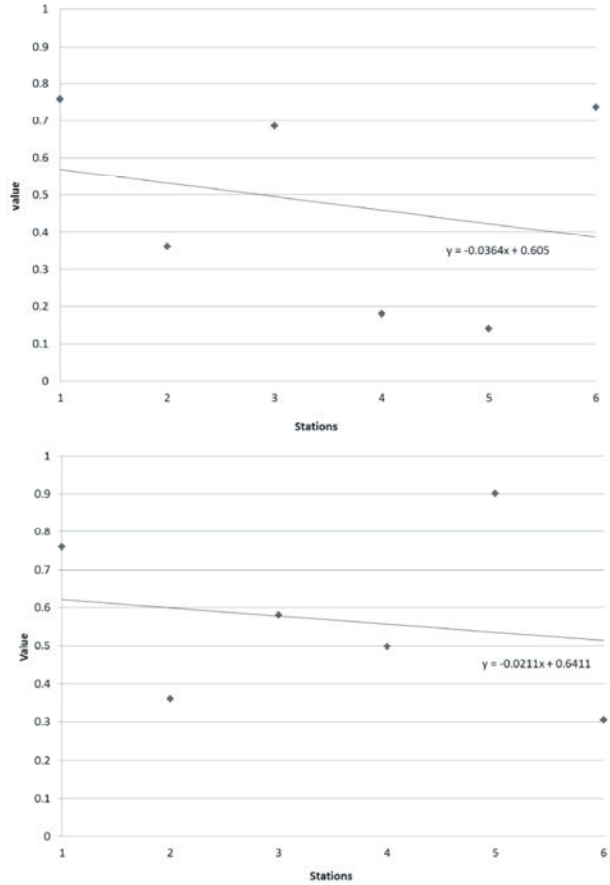

Fig. 8: CPI values for Darau River (down) and Inanam River (up)

In general, the concentrations of oil input from anthropogenic sources are increasing towards the estuary. The value of R2 is higher in Darau River rather than those observed in Inanam River. This presentation in Darau River is possibly related to the gradual input of hydrocarbons while in Inanam River a sudden increase was observed in Station 6 due to previously mentioned entrapment.

Carbon Preference Index (CPI): Carbon Preference Index (CPI) was applied in this research by applying the ratio of odd over even carbon number. The values of CPI in several calculations are based on the range of odd where compared to even carbon numbers. In this study the ranges of C_{25} to C_{33} were used [11, 13, 14, 34, 35]. The current CPI calculation follows the below equation.

$$\text{CPI } 25\text{-}33 = 0.5*[(C25\text{-}C33)/(C24\text{-}C32)] + [(C25\text{-}C33)/(C26\text{-}C34)] \quad (1)$$

As stated in equation 1, bigger CPI values indicate natural input where lower values may indicate anthropogenic sources. It is reported that CPI value close to one is sourced by recycled organic matters and/or marine microorganisms [14] as well as petroleum [11, 36, 34] where usually have no correlation to water quality parameter [37]. Predominant of vascular plants input to the environment usually demonstrate the CPI values from 3 to 6 [12, 13]. The results show that the CPI value is generally low since the proportion of natural input is lower than anthropogenic input. Examination of crude and processed oil reveals the same proportion of odd and even carbon numbers where CPI fluctuates around the unity. Dominant of the natural inputs in environmental samples increases the CPI value higher than unity. The reverse condition appears for anthropogenic hydrocarbon where the value decreases. The results of CPI values are presented in Figure 8. In comparison, Inanam River shows a range of CPI fluctuating from 0.3 to 0.9 while Darau River reveals lower

values ranging from 0.14 to 0.75 in various stations. This is noticeable that both rivers receive amounts of anthropogenic input from various sources however Inanam River is apparently more influenced by pollutants than Darau.

In conclusion, this study showed that untreated urban discharges materials transfer tremendous amount of organic pollutants to the aquatic ecosystems such as rivers [38]. Fingerprinting of environmental samples by application of molecular markers of aliphatic hydrocarbon presented in this paper demonstrated that the fingerprinting is an approachable technique to evaluate and monitor changes of sediment chemistry. The application of the method would help researchers and decision makers of the country not only to estimate the pollution loads but also to monitor the environmental quality as well as evaluating the rehabilitation processes of threatened ecosystems within the human settlements. Since the application of the technique is quite new, the authors suggest further studies to overcome the weaknesses of the method and improve the accuracy and appropriateness.

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