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ORIGINAL PAPER

Seasonal variations in the distribution of aliphatic hydrocarbons in surface sediments from the Selangor River, Peninsular Malaysia's West Coast

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Abstract The seasonal variation of petroleum pollution including *n*-alkanes in surface sediments of the Selangor River in Malaysia during all four climatic seasons was investigated using GC–MS. The concentrations of *n*-alkanes in the sediment samples did not significantly correlate with TOC ($r = 0.34$, $p > 0.05$). The concentrations of the 29 *n*-alkanes in the Selangor River ranged from 967 to 3711 µg g^{-1} dw, with higher concentrations detected during the dry season. The overall mean per cent of grain-sized particles in the Selangor River was $85.9 \pm 2.85\%$ sand, $13.5 \pm 2.8\%$ clay, and $0.59 \pm 0.34\%$ gravel, respectively. *n*-alkanes are derived from a variety of sources, including fresh oil, terrestrial plants, and heavy/degraded oil in estuaries. The results of this study highlight concerns and serve as a warning that hydrocarbon contamination is affecting human health. As a result, constant monitoring and assessment of aliphatic hydrocarbons in coastal and riverine environments are needed.

Keywords n-alkane · Sediments · Selangor River · Degradation · Pollution

Introduction

The majority of petroleum hydrocarbons, which are regarded as priority pollutants and biohazards in the ecosystem, consist of *n*-alkanes, which are harmful

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to both human health and the environment. *n*-alkanes are saturated hydrocarbons, meaning that only single bonds between hydrogen and carbon are present in them. The formula for *n*-alkanes, C_nH_{2n+2}, demonstrates the strongest possible bonds between the atoms of carbon and hydrogen. Although petroleum is the main source of *n*-alkanes globally, they are naturally occurring substances. Alkanes can be solids, liquids, or gases at room temperature. Methane, ethane, propane, and butane are gases, the unbranched alkanes pentane through hexadecane are liquids, and the homologs larger than hexadecane are solids (Honda & Suzuki, 2020; Patel et al., 2020).

Petroleum pollution has long been a serious problem due to their carcinogenic and mutagenic impacts. Unintentional oil spills and rising global demand for gasoline are two of significant causes of petroleum discharge into the environment (Retnam et al., 2013; Singh et al., 2012). For example, *n*-alkanes are one form of chemical that makes up petroleum hydrocarbons.

The Strait of Malacca is a narrow waterway that runs along the west coast of Peninsular Malaysia. Due to the large number of oil tankers that pass through it, both routine tanker operations activities such ballast water discharges and unintended oil spills are more frequent (Masood et al., 2018). The Malaysian Marine Department stated in 2003 that 127 oil spill incidents have occurred in the Malacca Strait since 1976 as a result of high oil tanker traffic (Rusli, 2011).

Since the 1980s, tens of thousands of hydrocarbon pollution investigations have been conducted to determine the petroleum hydrocarbon contamination in the air, sediment, biota, and water (Alkhadher et al., 2023a, b, c, d, e, f, g; Sharifi et al., 2022; Wang & Stout, 2007). These studies found elevated levels of hydrocarbons such as polycyclic aromatic hydrocarbons (PAHs), linear alkylbenzenes (LABs) and *n*-alkanes in a variety of developed, industrialized, and urbanized regions, as well as busy, heavily visited maritime ports and harbour points. The majority of these research have been carried out mostly in northern hemisphere countries with seasonal fluctuations, such as the United States of America, Japan, England, Norway, and Canada (Wang & Stout, 2007).

An assessment of studies on hydrocarbon pollution reveals a sizable gap in investigations of hydrocarbon pollutants in tropical nations like Malaysia, and this investigation is a part of a long-term hydrocarbon monitoring effort in Malaysia (Zakaria et al., 2002; Boonyatumanond et al., 2006; Saha et al., 2009; Mirsadeghi, 2010; Vaezzadeh et al., 2014, 2015a, 2015b, 2017a, 2017b; Masood et al., 2016; Keshavarzifard et al., 2020; Sharifi et al., 2022; Alkhadher et al., 2023e, f, g, Mohd Sallan et al., 2023; Roslee et al., 2023).

It is well recognized that *n*-alkanes in estuarine and marine environments come from both biological and human-made sources (Keshavarzifard et al., 2020; Yu et al., 2019). Aliphatic hydrocarbons (AHs) are found in sediment from a variety of sources, such as anthropogenic sources, higher terrestrial plants, and petroleum wastes, as well as autochthonous sources, which are primarily derived from higher aquatic species like algae, microorganisms, and aquatic macrophytes (Keshavarzifard et al., 2020; Liu et al., 2012; Sharifi et al., 2022). Shipping oil, tanker operations such as cleaning and deballasting, crankcase oil use, and fishing are all significant man-made sources of petrogenic *n*-alkanes. Selective oxidation, biodegradation, chemical reactions, evaporation, and photooxidation can all change the composition of hydrocarbons (Keshavarzifard et al., 2020; Masood et al., 2021).

The information available on potential (mixed) halogenated *n*-alkanes (PXAs) emphasizes the critical need to identify and monitor these potential pollutants, preferably using mass spectrometry. It is calculated that 60,000 pixels will have enough resolving ability to distinguish 74% of the priority elemental compositions from the most likely interferents, which are chlorinated alkanes (full width at half-maximum), implying that the majority of high-resolution mass spectrometers will be able to accurately identify the PXAs (Li et al., 2021). Petroleum biomarkers, which include isoprenoid *n*-alkanes, PAHs, and hopanes, have a direct link to biological precursors (Mici c et al., 2013; Peters et al., 2005).

In order to evaluate hydrocarbon inputs from anthropogenic sources, the Selangor River, the largest river on Malaysia's west coast, conducted a rigorous research of compound-specific hydrocarbon pollution. The seasonal trend of aliphatic hydrocarbons has never been thoroughly studied in Malaysia previously. The main objective is to determine the source of aliphatic hydrocarbons present in the sediments of the Selangor River and its tributaries as well as measure their distribution and concentration.

Methodology

Sampling

Study area

The sediments were collected from the Selangor River, which is on Peninsular Malaysia's west coast and faces the Straits of Malacca. The number of ships and tankers that use the narrow Straits of Malacca, which connect Sumatra, Indonesia, with Peninsular Malaysia, makes it one of the busiest waterways worldwide (Abdullah et al., 1999; Thia-Eng et al., 2000). The surrounding districts of Selangor state are also experiencing development, and the state's natural resources are becoming increasingly stressed. In the research area, there are still pockets of upland forest, marsh forest, and some mangroves. Rubber plantations in the area are rapidly becoming more viable urban areas or oil palm farms as a result of the decline in worldwide market prices brought on by the advent of synthetic rubber. The predominant land use in the study region is agriculture, especially the oil palm, paddy, and coconut plantations (Fig. 1). In addition, it provides challenging recreational pursuits for tourists who are adventurous, ecotourists, and travellers such as river tubing, whitewater rafting, and kayaking.

Sampling locations

In this study, thirteen stations with the codes S1–S13 were chosen for sediment sample collection in 2018 (Fig. 1). The Global Positioning System (GPS) was used to pinpoint the exact location of the sampling points and stations, and sample sites were selected based on the presence of anthropogenic activities and potential hydrocarbon sources. Since the sample is expected to have different levels of pollutants throughout time, sediment samples were collected during the rainy inter-monsoonal period, the dry inter-monsoonal period, the southwest monsoon (SWM) period, and the northeast monsoon (NEM) period. Furthermore, duplicate analysis was carried out for reproducibility and a coefficient efficient for AH was below 10%. Table 1 summarizes the sampling locations and the application of the Ekman dredge sampler to collect surface sediment samples. The top 5-cm layer of the surface sediment was carefully sub-sampled at each sampling location using a metallic spatula. After being immediately transferred to polyethylene bags with zip-ties, the samples were then brought to the lab in a cooler box where they were kept in a freezer at a temperature of –20 °C for additional experiments.

Chemical analysis

The samples were extracted, purified, and fractionated using a previously reported method (Zakaria et al., 2002; Masood et al., 2014; Magam et al., 2016; Alkhadher et al., 2015, 2016, 2020a, 2020b, 2020c). Dried sediment was pulverized in a porcelain mortar with a 2-mm mesh sieve. 250 mL of dichloromethane (DCM) was used in a Soxhlet system to extract the hydrocarbons, and the bottom round flasks were used to collect the extracts, which were then treated with copper granules to eliminate the sulphur, which is known to interfere with GC analysis. The extract volume was reduced by rotary evaporation before being loaded onto a glass chromatographic column containing silica gel activated with 5% H_2O. The elution solvent for the hydrocarbon fraction was 20 mL Hexane/DCM (3:1, v/v). The extracts were rotary evaporated to 1–2 mL before being separated gradually on a silica gel column that had been fully activated with 4 mL of high-grade hexane. The fraction of aliphatic hydrocarbons from the second step column chromatography was rotary evaporated to approximately 1 ml and then transferred to a 1.5 mL amber vial. The 1 mL sample was carefully reduced to near dryness using a stream of nitrogen gas and then redissolved in 100 µL of isooctane.

In a GC (7890A) coupled to a C5975 quadrupole mass-selective detector (SIM mode), a 30-m fused-silica capillary column (HP-5MS) is employed for n-alkane analysis. The carrier gas used was pure helium. The electron ionization (EI) method was used for MS at a 70-eV energy level, 200 °C for the source temperature, and ~ 2000 eV for the electron multiplier voltage. Samples were injected splitlessly into the GC injection port at a temperature of 300 °C, then a 1-min purge after that. The temperature of the column increased from 70 °C for 1-min to 150 °C at a rate of 30 °C per minute, then 5 °C per minute to 310 °C, where it remained for 10 -min. The *n*-alkanes were monitored at the following *m/z* values: *142 (C_{10}), 170 (C_{12}), 198 (C_{14}), 254 (C_{18}), 282 (C_{20}), 310 (C_{22}), 338 (C_{24}), 366 (C_{26}), 394 (C_{28}), 422 (C_{30}), 450 (C_{32}), 478*

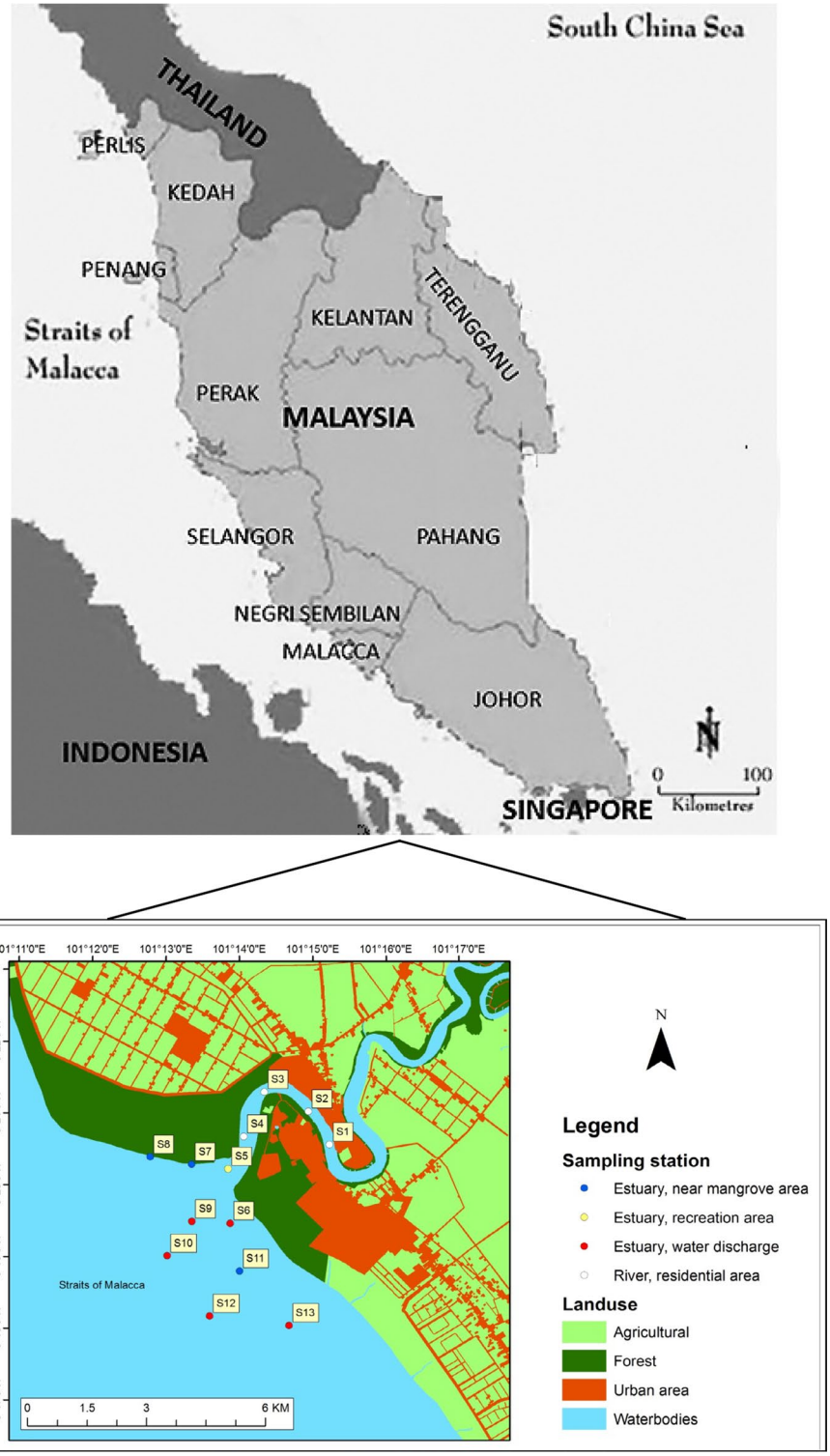

Fig. 1 Locations of sampling stations in Selangor River, Malaysia

(C_{34}), 506 (C_{36}), 534 (C_{38}), and 562 (C_{62}) (C_{40}). A 191 *m/z* ion monitoring technique was used.

Every set of samples was injected with one of five distinct external standards for *n*-alkanes, even those

Table 1 Sampling locations and description

Station (%)	TOC		Geographical coordination		Description of
	Rainy	Dry	North	East	
S1	1.32	0.83	03° 20′ 10''	101° 15′ 25''	River, residential area
S2	1.25	1.36	03° 20′ 30''	101° 15′ 05''	River, residential area
S3	2.74	1.52	03° 20′ 45''	101° 14′ 30''	River, residential area
S4	1.28	1.09	03° 20′ 10''	101° 14′ 15''	River, residential area
S5	1.35	1.41	03° 19′ 48''	101° 14′ 08''	Estuary, recreation area
S6	1.45	1.30	03° 19′ 10''	101° 14′ 10''	Estuary, waste discharge
S7	1.51	0.87	03° 19′ 50''	101° 13′ 40''	Estuary, near mangrove area
S8	1.08	1.34	03° 19′ 55''	101° 13′ 05''	Estuary, near mangrove area
S9	1.89	0.84	03° 19′ 10''	101° 13′ 48''	Estuary, waste discharge
S10	0.92	1.24	03° 18′ 45''	101° 13′ 25''	Estuary, waste discharge
S11	1.31	1.05	03° 18′ 40''	101° 14′ 15''	Estuary, near mangrove area
S12	1.18	1.25	03° 18′ 05''	101° 13′ 55''	Estuary, waste discharge
S13	1.14	1.06	03° 18′ 00''	101° 14′ 55''	Estuary, waste discharge

with a low carbon number (C_{10}–C_{36}). By comparing the target analytes' mass to charge ratio (*m*/*z*) and retention times to standards, the target n-alkane was recognized. The contents were determined by comparing the integrated sample peak area to the standard peak area.

Quality control and quality assurance

QC and QA procedures were carried out during the experimental analysis of this study to ensure the certainty and accuracy of the analytic method. The surrogate recovery for sediment samples was within the acceptable range (80–120%). Even number *n*-alkanes made up the external standards for *n*-alkanes with $C_{10–40}$. Daily preparation of fresh standards was carried out for the analysis. Each batch of set samples was subjected to a blank procedure, and the analysis of the reagent blanks showed that the analysis apparatus and glassware were free of contamination. Additionally, after each batch of samples was analysed using an instrument, a purge was processed.

Total organic carbon (TOC)

The method presented via Bakhtiari et al. (2010) and Raza et al. (2013) is the analytical process that was used to evaluate the TOC content. The sediment samples were mashed and homogenized using a mortar and pestle after being dried in an oven for an entire night at 60 °C. Each sediment sample was weighed to determine its mass, and then, 1–2 mL of a 1M hydrochloric acid (HCl) solution was put to completely dissolve any inorganic carbon. Samples were then dried at 100 °C for 10 h to remove the HCl. Each sample was reweighed to a weight of about 1 g, and the TOC was then measured using an LECO CR-412 carbon analyser at 1350 °C for 60s. TOC is expressed for this study as mg g^{-1}.

Grain size analysis

Granulometry is the measurement of the sediment grain size of a granular substance as a proportion of the weight of various sized particles. The four procedures that were used to the sediment samples that were obtained for this investigation were washing, keeping in a low-temperature oven, disaggregating, and stiliting. Moreover, almost 50 g of each sample was sieved for 20 min through a standard set of sieves arranged in a one phi class interval. According to Moopam (1999), the categories of sediment size that were used in this study are as follows: granule (gravel) is > 2000 m, very coarse sand is > 1000 m, coarse sand is > 500 m, medium sand is > 250 m, fine sand is > 125 m, very fine sand is > 62 m, and clay is 63 m.

Statistical analysis

Data were statistically analysed using IBM-SPSS software, version 21.0. To assess the variations in

concentrations across the sampling locations, analysis of variance (ANOVA) was used to detect the meaningful differences in the levels of *n*-alkanes between the sampling stations and between the four seasons. The difference in n-alkane concentration between the sampling sites under comparison was deemed statistically significant when $p < 0.05$. Using Spearman's rank correlation analysis, the correlations between the hydrocarbons (HCs) and TOC in the sediments were examined (Zhang et al., 2006). To identify likely n-alkane linear combination that would be useful to distinguish their sources, the PCA was used. With the factor loadings obtained from PCA, clustering analysis was done to eliminate any correlations between the variables. Using the eigenvalue > 1 criteria, the principal components were retrieved following varimax rotation (Dickhut et al., 2000; Liu et al., 2009; Sharifi et al., 2022; Simpson et al., 1998).

Results and discussion

n-alkane concentration in the Selangor River's sediment

The concentration and distribution of *n*-alkanes (nC_{10}–nC_{36}) in sediments from the Selangor River were investigated. Table 2 indicated that the total concentrations of *n*-alkanes ranged from 967 to 3711μg g^{-1}dw (Fig. 2). The compositional characteristics of *n*-alkanes were found to be largely constant across all locations. As shown in Fig. 3, the sediments from S9 at downstream of the River had the highest concentration of *n*-alkanes with 3711 μg g^{-1}dw), whereas the sediments from S3 at upstream of the river had the lowest concentration with 967 μg g^{-1}dw. The variation in n-alkane concentrations at riverine and estuarine stations may result from both anthropogenic sources, such as shipping activities, industrial, domestic waste discharges, and input from the environment, such as microbial activities, submerged or floating macrophytes, and emerging terrestrial plant (Li, et al., 2021; Oyo-Ita et al., 2010; Sharifi et al., 2022).

The station S2, which is near the waste disposal plant for Selangor City, has the second-highest overall n-alkane level (3498 μg g^{-1}dw). This shows that the direct discharge of domestic wastewater, which occurs close to this station, is a significant source of *n*-alkanes in the riverine sediment. The area around station S2 is protected from the sea's waves, particularly those from the Strait of Malacca. This probably reduces water flow and increases sedimentation, causing the deposition of the *n*-alkanes related to the suspended particles that are transported down the river, according to Zheng et al. (2002).

Overall, it was found that the upstream areas of the Selangor River contained higher levels of *n*-alkanes than the downstream parts by comparing the n-alkane content in sediment samples. This result may be explained by the fact that there are more habitations and industrial facilities in the Selangor River's upstream than in its downstream. The concentration of total *n*-alkanes, on the other hand, decreased from the dry to the wet season, most likely as a result of the *n*-alkanes being diluted by the increased rainfall and river flow during the wet seasons. The outcomes of the one-way ANOVA indicated that the differences between the *n*-alkanes specifically C_{19}, C_{20}, C_{23}, and C_{24} among the 13 sampling stations were significant ($p < 0.05$; Table). This can be ascribed to diverse human activity at each station, which may result in distinct emission sources. Meanwhile, the concentrations of most *n*-alkanes examined in surface sediment differed significantly between the four seasons ($p < 0.05$), indicating that temporal fluctuation had a large influence on n-alkane concentrations. Additionally, the results showed that C_{34} predominated, which is consistent with the mixed n-alkane origins of the sediments from the Selangor River. They also showed that C_{17}–C_{34} predominated during the dry intermonsoonal period, C_{34}–C_{36} predominated during the SWM period, and C_{36} and C_{26}–C_{34} predominated during the NEM period. These results of the study are referring to show different carbon chain length predominance in different monsoon seasons. There are some possible explanations for this trend, which include the differences in environmental conditions such as temperature and salinity or the differences in microbial degradation rates while hydrocarbons can be degraded by microbes, and the rate of degradation can vary depending on the type of hydrocarbon and the environmental conditions (Gayantha et al., 2021).

The LMW/HMW *n*-alkanes (L/H) values for stations S2, S3, S5, S7, S9, and S10 were less than 1, as shown in Table 2. L/H-alkane ratios in other sediment samples that ranged from 1.07 to 1.46 were larger than one. The LMW *n*-alkanes may have undergone

Table 2 Average concentrations of *n*-alkanes (μg g^{-1}dw) during the four studied periods (Dry, ---) in the surface sediments of the Selangor River

Compound	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12	S13	Sig (*p* value)	
														Among stations	Among seasons
10	121	88	114	119	179	169	142	162	205	92	159	178	127	0.65	0.000
11	65	64	62	64	105	105	75	87	280	75	84	105	98	0.39	0.014
12	31	48	30	29	60	66	34	40	326	65	38	61	80	0.27	0.116
13	22	37	21	40	54	55	24	45	217	60	37	60	68	0.42	0.001
14	8	20	8	57	46	40	8	53	50	51	35	58	50	0.995	0.000
15	6	10	7	30	24	22	6	32	28	27	20	29	30	0.997	0.000
16	5	0	7	4	4	5	5	11	6	4	6	0	11	0.874	0.000
17	38	66	29	29	40	44	31	31	24	59	15	45	31	0.348	0.042
Pristane	33	19	13	13	11	15	14	12	12	15	13	10	8	0.317	0.014
18	63	116	46	48	68	73	50	46	37	101	22	79	45	0.22	0.056
Phytane	16	21	17	8	20	14	13	15	11	17	13	33	11	0.489	0.067
19	53	83	27	33	70	79	48	65	44	101	23	68	44	0.029	0.410
20	40	42	3	14	72	86	46	88	52	101	23	55	43	0.043	0.485
21	35	40	6	16	45	56	39	74	49	63	25	40	41	0.018	0.548
22	29	39	9	17	16	25	32	60	45	23	27	24	39	0.276	0.002
23	52	37	15	35	56	52	64	79	56	21	39	41	52	0.026	0.043
24	80	34	23	57	104	83	101	101	68	19	53	61	67	0.003	0.530
25	95	166	23	60	109	88	120	112	85	82	64	71	71	0.376	0.385
26	116	352	24	64	116	95	147	127	109	170	78	87	77	0.4	0.569
27	87	260	21	46	88	58	125	90	90	138	58	67	55	0.263	0.433
28	62	185	19	30	65	28	107	59	74	111	42	52	37	0.154	0.285
29	56	197	19	34	92	57	88	77	122	134	61	71	65	0.119	0.261
30	51	206	19	37	112	80	73	90	159	151	76	85	86	0.241	0.306
31	45	143	41	54	94	62	77	113	165	127	57	58	59	0.24	0.009
32	35	36	79	82	62	32	83	154	176	87	25	12	13	0.216	0.001
33	57	75	63	75	201	104	131	173	323	123	72	116	84	0.281	0.000
34	88	131	40	64	402	207	201	201	535	174	141	267	186	0.235	0.003
35	70	427	82	40	190	139	125	91	244	132	78	137	166	0.55	0.624
36	63	556	100	30	97	109	92	43	117	113	51	81	158	0.506	0.502
[a]∑*n*-alkanes	1521	3498	967	1230	2601	2048	2099	2332	3711	2437	1436	2052	1903	–	–
[b]C31/C19	0.86	1.73	1.55	1.65	1.35	0.79	1.59	1.75	3.77	1.26	2.52	0.85	1.34	–	–
[c]CPI	0.97	0.98	0.97	0.98	1.02	1.00	0.97	0.98	1.04	1.00	1.00	1.03	1.01	–	–

Table 2 (continued)

Compound	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12	S13	Sig (*p* value)	
														Among stations	Among seasons
[d]ACL	28.31	28.29	30.20	29.28	29.65	29.19	28.90	29.52	30.40	29.24	29.11	29.42	29.18	–	–
[e]C17/Pri	1.15	3.55	2.25	2.18	3.56	2.95	2.27	2.68	1.93	3.85	1.15	4.34	3.76	–	–
[f]C18/Phy	4.04	5.48	2.65	5.68	3.46	5.16	3.91	2.97	3.42	5.99	1.65	2.43	4.10	–	–
[g]Pri/Phy	2.10	0.88	0.75	1.57	0.58	1.05	1.05	0.75	1.12	0.91	0.96	0.32	0.74	–	–
[h]ΣC_{even} *n*-alkanes	792	1853	520	653	1402	1098	1120	1236	1961	1263	776	1099	1019	–	–
[i]ΣC_{odd} *n*-alkanes	681	1605	417	555	1168	921	952	1069	1727	1142	633	909	864	–	–
[j]Odd/Even	0.86	0.87	0.80	0.85	0.83	0.84	0.85	0.86	0.88	0.90	0.82	0.83	0.85	–	–
[k]L/H	1.40	0.56	0.94	1.46	0.83	1.30	0.88	1.11	0.84	0.86	1.13	1.12	1.07	–	–
[l]TAR	1.95	3.78	1.29	1.46	2.04	1.23	3.38	2.20	3.97	2.13	3.05	1.38	1.71	–	–
TOC (%)	1.22	1.36	1.99	1.30	1.37	1.45	1.85	1.21	1.10	1.13	1.30	1.40	1.30	–	–

[a]$\sum n$ alkanes = Sum of the concentrations of the C_{10} to C_{36} alkanes; [b]C_{31}/C_{19} = Ratio of C_{31} alkane to C_{19} alkane;[c]CPI = Carbon preference indices; [d]ACL = Average chain length value of odd carbon numbers from C_{25} to C_{33}; [e]C_{17}/Pr = Ratio of C_{17} n-alkane to pristane, [f]C_{18}/Ph = Ratio of C_{18} n-alkane to phytane, [g]Pr/Ph = Ratio of pristane over phytane;[h]$\sum$Even Carbons = Sum of the concentration of C_{10}, C_{12}, C_{14}, C_{16}, C_{18}, C_{20}, C_{22}, C_{24}, C_{26}, C_{28}, C_{30}, C_{32}, C_{34}, and C_{36}; [i]$\sum$Odd Carbons = Sum of the concentrations of C_{11}, C_{13}, C_{15}, C_{17}, C_{19}, C_{21}, C_{23}, C_{25}, C_{27}, C_{29}, C_{31}, C_{33}, and C_{35}; [j]Odd/Even = The ratio of even carbon alkanes to odd carbon alkanes; [k]L/H = The ratio of low molecular weight alkanes to high molecular weight alkanes; [l]TAR = Terrestrial to aquatic ratio (sum of the concentrations of C_{27}, C_{29}, and C_{31} alkanes to sum of concentrations of C_{15}, C_{17}, and C_{19} alkanes)

more microbial degradation than the HMW *n*-alkanes in samples with low L/H *n*-alkanes, similar to a study conducted in the Peel Plateau in northwestern Canada (Bröder et al., 2021). Furthermore, LMW hydrocarbons may also be subject to greater degradation by living organisms due to their increased bioavailability (Gaspare et al., 2009; Keshavarzifard et al., 2017). Consequently, LMW biodegradation could be the cause of reduced LMW n-alkane concentrations at some of the stations under study (Keshavarzifard et al., 2020). The prevalence of HMW n-alkane with odd numbers of carbon atoms in certain of the sediment sample locations can be attributed to input of n-alkane from terrestrial high plants. Odd carbon atom alkanes often come from biogenic sources, while even carbon atom alkanes typically come from anthropogenic sources (Kang et al., 2018; Sharifi et al., 2022; Volkman et al., 1992).

High L/H-n-alkane ratios ranging from 0.01 to 13.32 were reported in a recent study of sediment in Malaysia's Prai Strait. The L/H n-alkane ratios discovered in the current study are lower than those mentioned in the previous study (Sakari et al., 2008a). Zakaria et al. (2002) found significant petrogenic hydrocarbon imports onto Peninsular Malaysia's west coast. The major hydrocarbon, the n-alkane with the highest concentration of carbon atoms, can also be used as a source indicator. The most common *n*-alkanes in our study have a number of carbon atoms centred around 34 (C_{34}) (Table 2). Furthermore, the prevalence of *n*-alkanes with LMW suggests new oil inputs. In the LMW *n*-alkanes detected in the sediments, even carbon atoms predominated over odd carbon atoms, indicating that anthropogenic sources of *n*-alkanes predominated over biogenic sources (Sakari et al., 2008b). Odd over even numbers of carbon atoms were more abundant in the upper areas of the Selangor River, indicating higher inputs of *n*-alkanes from terrestrial plant sources. However, as one moves down the river, the pattern shifts to a majority of even-numbered carbon *n*-alkanes, emphasizing inputs from petrogenic sources.

Unresolved complex mixture (UCM)

The unresolved complex mixture (UCM) is a complex mixture of hydrocarbons with branching and cyclic structures, as well as certain overlapping peaks which generate a "hump" and cannot be detected by

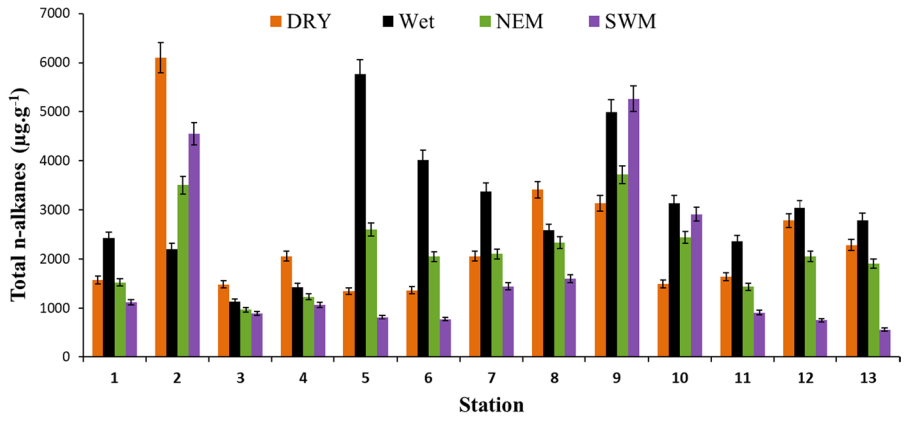

Fig. 2 Total concentrations of the *n*-alkanes in the sediment samples. Error bars indicate the 95% confidence interval

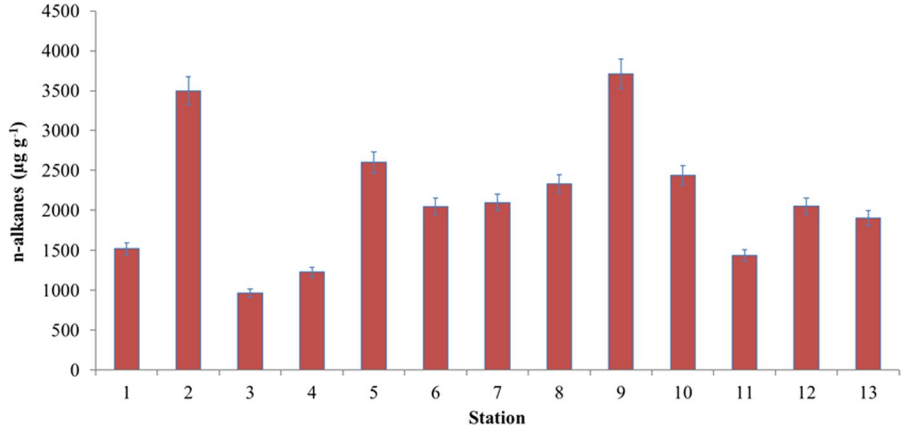

Fig. 3 Mean concentrations of total *n*-alkanes at each sediment sampling station. Error bars indicate the 95% confidence interval

a capillary GC column (Bouloubassi & Saliot, 1993; Bouloubassi et al., 2012). UCMs are a typical feature in environmental samples polluted with weathered petroleum residues (Gough & Rowland, 1990). Although UCMs are expected to be rather resistant to microbial degradation, they can impede the identification of alkanes (Gough et al., 1992; Hu et al., 2009). Although UCMs are mostly found in the HMW range, they can also be found in the LMW range and evaporate. A modest UCM in a typical chromatogram of *n*-alkanes implies short-term weathering. In the current study, chromatograms of *n*-alkanes showed UCMs in some of the samples. Figure 4 shows a typical chromatogram of *n*-alkanes with UCM hump (Retnam et al., 2013).

Carbon preference index (CPI)

CPI, as well as *n*-C_{17}/Pristane and *n*-C_{18}/Phytane ratios, was calculated to acquire a better understanding of the principal sources of hydrocarbons in sediment samples. According to Simoneit et al. (1991), the CPI is a measurement of the biologically produced *n*-alkanes that can be applied to determine the straight chain's degree geolipid diagenesis. In addition to being employed in organic geochemistry to denote the presence of geolipids with straight chains, it is a quantitative evaluation of the degree to which geological lipids retain the original biological chain length specificity (Meyers & Ishiwatari, 1995). Additionally, it compares the n-alkane contribution from anthropogenic (CPI 1) sources, such as PHCs or aquatic species, to those from natural (biogenic/terrestrial., CPI > 1) sources. The ratio of Codd *n*-alkanes/Ceven *n*-alkanes for the petrogenic is around 1.0 since the petroleum mixes also contain equivalent amounts of nearby members of hydrocarbons (Tran et al., 1997). According to values of this ratio (Table 2), petrogenic origin is the primary source of hydrocarbons in the research region.

According to formula proposed by Bakhtiari et al. (2011), Zhu et al. (2005), the CPI was estimated in the current study as follows:

$$\text{CPI is calculated as } \left(C_{23} + C_{25} + C_{27} + C_{29} + C_{31}\right)/\left(C_{24} + C_{26} + C_{28} + C_{30}\right) \quad (1)$$

The calculated CPI values are depicted in Fig. 5. The research area's average CPI values during the four study periods ranged from 0.97 to 1.04. Since the dominance of odd or even carbon atoms is missing, petrogenic sources are probable.

Along with the ratio Pr/Ph, other ratios, specifically C_{17}/pristane and C_{18}/phytane of the *n*-alkanes/acyclic isoprenoid *n*-alkanes, can be utilized to identify the sources of hydrocarbons in environmental ecosystem (Pavlova & Ivanova, 2003). Ratios of C_{17}/pristane and C_{18}/phytane levels below 1.0 may indicate weathered petroleum, whilst values above 1.0 are indicative of unpolluted environments. The ratios between C_{17} and C_{18} in pristane ranged from 1.15 to 4.34 and from 1.65 to 5.99, respectively (Fig. 5). Being greater than 1.0 at all sampling stations, values of these two ratios do not indicate presence of weathered petroleum in the sediments.

Since phytane commonly originates from petroleum, values of the pristane/phytane ratio less than 1.0 can indicate pollution with petroleum while values higher than 1.0 indicate biogenic inputs or bituminous coal (Readman et al., 2002; Volkman et al., 1992). For pristane and phytane, respectively, the oxidation and reduction of the chlorophyll's phytol side chains can be considered biogenic sources. These isoprenoids can also be found in the lipids of bacteria and zooplankton (Le Dréau et al., 1997). In the Selangor River, the pristane/phytane ratio ranged from 0.32 to 2.10 (Fig. 6). Nevertheless, majority of the sampling stations had values that were less than or close to 1.0, demonstrating the significance of petroleum imports in the studied area. In the bulk of the sampling stations, a falling pattern was seen in the pristane/phytane ratio values from the higher to the lower regions of the river. This pattern demonstrates once more that the river's upper portions have significant biogenic n-alkane sources, but the lower portions of the river have more significant petroleum hydrocarbon inputs.

Average chain length (ACL)

Average chain length (ACL) is another metric used to determine the environmental sources of *n*-alkanes and is based on the concentration of the odd carbon numbers of the *n*-alkanes in vascular higher plants (Boot et al., 2006; Poynter & Eglinton, 1990). With respect to plant species, latitude, and climatic conditions, the ACL is constant for places with a single source of *n*-alkanes, and however, petrogenic inputs cause the ACL to decrease in value. (Jeng, 2006). The formula for calculation of ACL is (Jeng, 2006):

$$\text{ACL} = \left(25 + C_{25} + 27 + C_{27} + 29 + C_{29} + 31 + C_{31} + 33\right)\left(C_{25} + C_{27} + C_{29} + C_{31} + C_{33}\right) \quad (2)$$

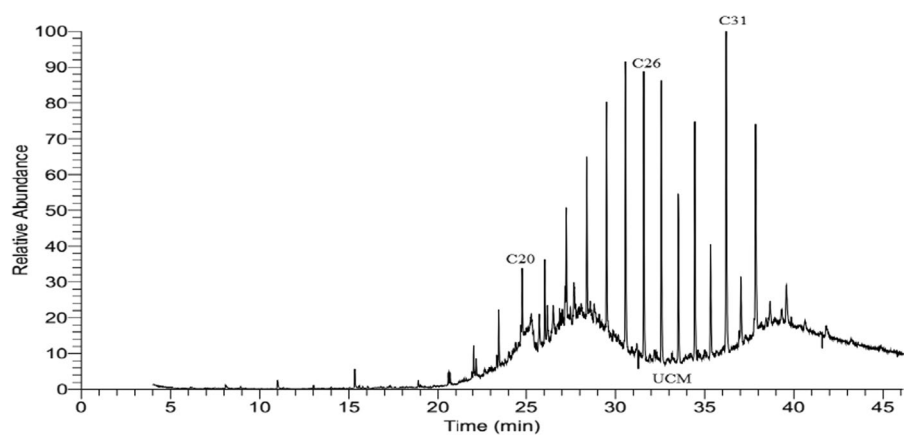

Fig. 4 A typical chromatogram of *n*-alkanes

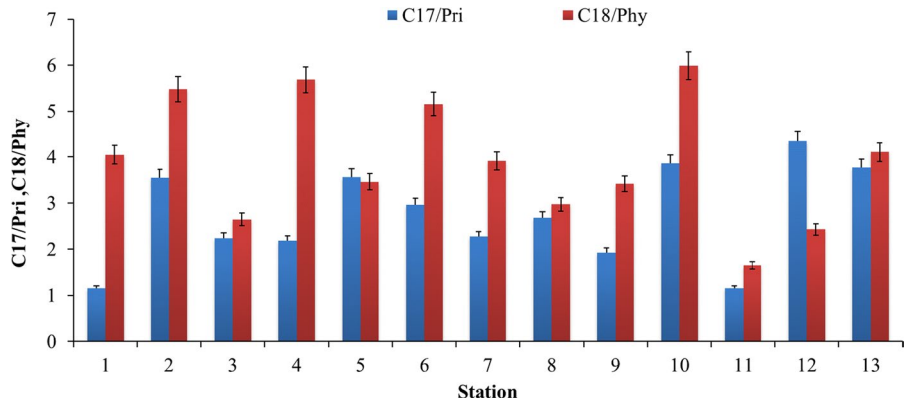

Fig. 5 Spatial variations of the n-C_{17}/pristane and *n*-C_{18}/phytane ratios. Error bars indicate the 95% confidence interval

The ACL results for the Selangor River sediments ranged from 28.29 to 30.40. This ACL value range may indicate the presence of petroleum inputs into the research area. In sediments from the Prai River, a wider range of ACL values (26.75–32.94) was recorded (Sakari et al., 2008b). This underlines higher petrogenic inputs in Prai River than the Selangor River, possibly due to occasional spillage of petroleum or due to other petrogenic inputs such as urban run-offs. Within this context, the very narrow ranges of ACL values for river sediments in the northeastern (29.6–29.8) and northwestern Taiwan (29.2–30.5 (Jeng (2006)) reveal lower petrogenic inputs in these areas.

Since *n*-alkanes from higher plants are associated to both the CPI and ACL, a correlation between them is anticipated (Jeng, 2006; Li et al., 2020). There was no correlation between the CPI and ACL values for the sediments in the current study ($P > 0.05$). This may be attributed to the fact that the *n*-alkanes used in calculations of the CPI and ACL did not originate from the same source of higher plants. According to Jeng (2006), when two (or more) *n*-alkanes have different ACL or CPI values, the ratio changes nonlinearly and disproportionally.

Terrigenous aquatic ratio (TAR)

Terrigenous aquatic ratio (TAR) compares sum of the concentrations of odd-numbered ($C_{27}+C_{29}+C_{31}$) *n*-alkanes, which are a characteristic of the debris of high terrestrial plant (Rieley et al., 1991), to the sum of the concentrations of odd-numbered ($C_{15}+C_{17}+C_{19}$) *n*-alkanes, which indicate the sources from aquatic environment

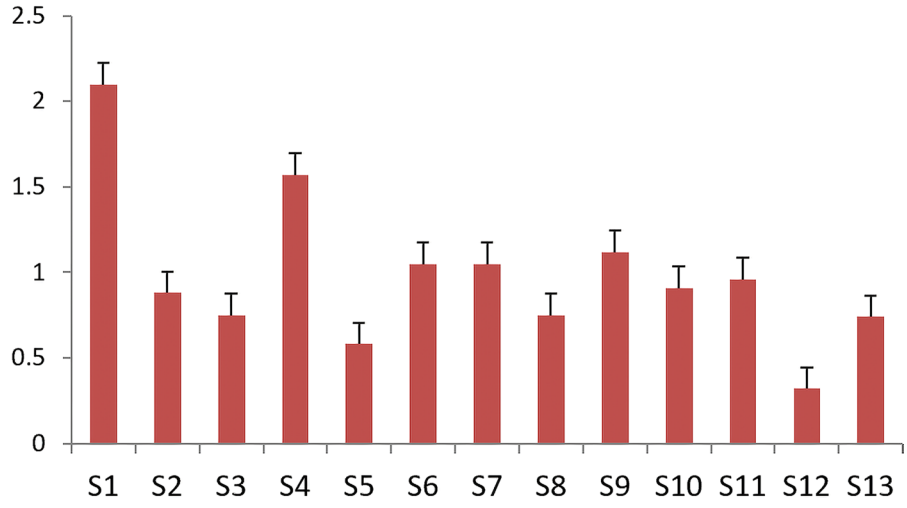

Fig. 6 Spatial patterns in the values of the pristane/phytane (Pr/Ph) ratio. Error bars indicate the 95% confidence interval

(Jaffe' et al., 2001). Therefore, the ratio of short-chain to long-chain n-alkane concentrations is measured by the TAR, which can be used to assess how significant terrestrial inputs are in comparison with aquatic ones (El Nemr et al., 2016). This is how the TAR is determined:

$$\mathrm{TAR} = \left(C_{27} + C_{29} + C_{31}\right) / \left(C_{15} + C_{17} + C_{19}\right) \quad (3)$$

When the TAR value is less than 1, it means that from aquatic sources, *n*-alkanes are more prevalent (Gomes & Azevedo, 2003). The findings of this study demonstrate that the TAR values for the sediment samples were > 1, which highlights that high terrestrial plant debris is a main source of the *n*-alkanes (Fig. 7).

Comparison of the levels of PAHs in sediments of Selangor River with those in other international literature areas

A number of previous researchers have reported on *n*-alkanes levels with the literature in the rivers around the world (Table 3). The comparison shows differences in the concentrations of the *n*-alkanes between Selangor River and the other locations worldwide. The average n-alkane concentration among stations calculated 2339 (μg g^{-1}). The finding from this study is significantly higher than previously reported by other scientists around the world (Iwegbue et al., 2021; Paletto et al., 2008) but consistent with findings from the east coast of Malaysia (Sakari et al., 2008a, 2008b), while it was less concentrated when compared with other regions (Ahad et al., 2011; Silva et al., 2012; Monza et al., 2013; Jeng, 2006; Sakari et al., 2008b).

Petroleum hydrocarbons, mainly consisting of *n*-alkanes and polycyclic aromatic hydrocarbons (PAHs), are considered as priority pollutants and biohazards in the environment, eventually affecting the ecosystem and human health (Liu et al., 2018). Furthermore, *n*-alkanes may eventually affect human health (Lu et al., 2012) and other social activities, such as agriculture (Zhang et al., 2017) and tourism (Zhu et al., 2015). Therefore, petroleum hydrocarbons are considered as priority pollutants (Ekperusi & Aigbodion, 2015), and its remediation is a matter of urgency. Generally, non-polluted area demonstrates hydrocarbons concentration (e.g. *n*-alkanes) less than 10 ng/mg (UNEP, 1992). The high concentrations of *n*-alkanes with their ratios showed that Selangor River, west Peninsular Malaysia, are heavily affected by anthropogenic pollutants. The results also highlighted that hydrocarbon pollutants would continue to be a problem, as long as there is a human population to produce it. In the coming years, the total amount of hydrocarbons discharged into the rivers and coastal waters in Peninsular Malaysia will progressively increase and add to the pollution already affecting parts of the region.

Correlation between total *n*-alkanes and TOC

Both terrestrial and marine sources contribute to the organic carbon (OC) found in sediments from aquatic environment (Prahl et al., 1994; Retnam et al., 2013).

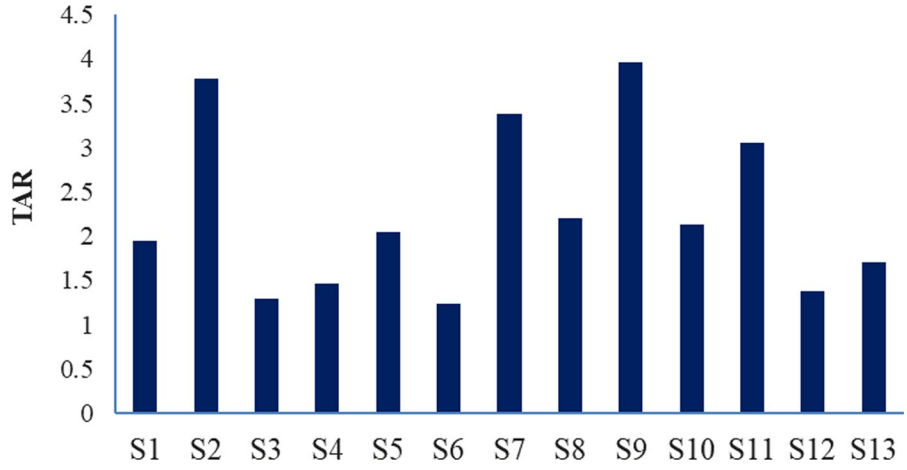

Fig. 7 Spatial pattern of the terrigenous aquatic ratio (TAR)

While the contributions of air deposition may be overlooked, fluvial transport is the primary process responsible for the input and accumulation of terrestrial total organic carbon TOC in the aquatic and marine sediments. Even carbon atom *n*-alkanes commonly originate from anthropogenic sources, while the source of odd carbon atom *n*-alkanes is biogenic (Kang et al., 2018; Retnam et al., 2013; Volkman et al., 1992).

The aliphatic hydrocarbons are substances that repel water. They have a propensity to sorb to organic materials once in an aquatic environment. The sediment's organic carbon content may have a significant impact on the quantities of hydrocarbons due to the sorptive tendency of aliphatic hydrocarbons. Peninsular Malaysia has a large number of plants; thus, the intense tropical rains can wash large quantities of organic materials out of the soil and into the rivers. At the same time, a significant amount of organic matter is known to be present in the mangrove environment.

In the Selangor River' sediments, the TOC concentration varied between 1.16 and 1.98%. Table 4 shows that there was no correlation between TOC and n-alkane levels that was positive ($r = 0.336$, $p > 0.05$). The fact that TOC and hydrocarbon concentrations have separate sources may be one factor in their lack of association. While the research area's aliphatic hydrocarbons primarily resulted from anthropogenic inputs, the sediments' main sources of TOC are often naturals (Mirsadeghi et al., 2013; Ouyang et al., 2006; Schumacher, 2002). Other investigations (Cavalcante et al., 2009; Mirsadeghi et al., 2013; Tam et al., 2001) revealed there was no correlation between TOC and hydrocarbon contents, whilst other studies suggested a connection between TOC and hydrocarbon concentrations (Bush & McInerney, 2015; Chen et al., 2012; Huang et al., 2012). Given this, further information is required to completely comprehend the interactions between TOC and hydrocarbon concentrations in the tropical region.

Table 3 The concentrations (μg g^{-1}) and indices of *n*-alkanes in the literature rivers around the world

n-Alkane index	Location							
	Argentina (Marine) Paletto et al. (2008)	Tyne, UK Ahad et al. (2011)	Alagoas, Brazil Silva et al. (2012)	Argentina (River) Monza et al. (2013)	Taiwan (Marine) Jeng (2006)	Penang, Malaysia Sakari et al. (2008b)	Escravos (River) Iwegbue et al. (2021)	This study (Selangor River) Masood et al. (2021)
[a]CPI	0.44–1.97	1.8–6.4	2.9–6.8	1.16–14.36	1.43–1.97	0.00–1.22	0.24–3.46	0.97–1.03
[b]ACL	–	28.5–29.5	28.5–29.4	–	28.4–29.3	26.75–32.94	25.5–30.8	28.29–30.40
[c]MH	C_{17}	C_{31}	C_{31} & C_{18}	C_{29}	C_{17}	C_{18} & C_{20}	C17	C34
[d]L/H	1.11–4.99	[i]nd	nd	0.04–1.19	~1	0.01–13.32	2.9–67.6	1.07–1.46
[e]Pr/Ph	0.04–1.19	3.48–7.86	nd	0.19–1.12	0.95–3.46	[i]nd	0.04–1.10	0.32–2.10
[f]C17/Pr	0.7–5.3	0.14–0.42	nd	0.2–6.95	0.97–1.21	nd	1.90–44.7	1.15–4.34
[g]C18/Ph	1.22–4.41	nd	nd	0.71–5.24	2.53–4.45	nd	0.03–6.24	1.65–5.99
[h]$\sum$*n*-alkanes	0.20–1.100	6.500–26.600	35–20.600	0.410–54.050	1.140–7.140	421–10,770	95–3430	967–3711
[j]UNEP Guideline	lower	higher	higher	higher	lower	higher	lower	lower

[a]Overall CPI = The carbon preference index $(C_{15}\text{-}C_{35}) = ½ \times [(C_{15}+C_{17}+C_{19}+C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33}) + (C_{17}+C_{19}+C_{21}+C_{23}+C_{25}+C_{27}+C_{29}+C_{31}+C_{33}+C_{35}) \div (C_{16}+C_{18}+C_{20}+C_{22}+C_{24}+C_{26}+C_{28}+C_{30}+C_{32}+C_{34})]$, [b]ALC = Average carbon chain value of odd carbon numbers from C_{25} to $C_{33} = [(25 \times (C_{25}) + 27 \times (C_{27}) + 29 \times (C_{29}) + 31 \times (C_{31}) + 33 \times (C_{33})) \div (C_{25}+C_{27}+C_{29}+C_{31}+C_{33})]$, [c]MH = Maximum n-alkane carbon number, [d]L/H = Sum of LMW *n*-alkanes over sum of HMW *n*-alkanes, [e]Pr/Ph = Ratio of pristane over phytane, [f]C_{17}/Pr = Ratio of C_{17} n-alkane over pristane, [g]C_{18}/P = Ratio of C_{18} n-alkane over phytane, [h]$\sum$ *n*-alkanes = Sum of aliphatic hydrocarbons between C_{10}—C_{36}, [i]nd = Not detected, [j]The concentration index limit of hydrocarbons (< 10,000 ng/g) in the environment, (UNEP, 1992)

Table 4 Correlation between the total concentration of *n*-alkanes and TOC

	n-alkanes	TOC (%)	
n-alkanes	Spearman Correlation	1	−0.360
	Sig. (2-tailed)		0.227
	N	13	13
TOC (%)	Spearman Correlation	−0.360	1
	Sig. (2-tailed)	0.227	
	N	13	13

Grain-size distribution

Grains have been divided into three groups based on their sizes: gravel, sand, and clay. Table 5 displays the particle size distribution of the sediments taken from the Selangor River. Study results revealed very high variation in the grain sizes of the sample sediments. The distributions of the grain sizes differed from a location to another in the study area. However, in the Selangor River sediments, the sandy fraction predominated, and the amount of clay in all the layers was larger than that of gravel. The average percentages of sand, clay, and gravel were 85.96 ± 2.85, 13.45 ± 2.80, and $0.59 \pm 0.34\%$, respectively.

Table 5 Grain size distributions in the sediments of the study area

Station	Gravel (%)	Sand (%)	Clay (%)
S1	0.89	80.20	18.91
S2	0.46	87.23	12.31
S3	0.86	88.89	10.25
S4	0.34	85.25	14.41
S5	0.56	85.02	14.42
S6	1.42	88.36	10.22
S7	0.45	84.50	15.05
S8	0.65	82.72	16.63
S9	0.35	89.01	10.64
S10	0.51	85.03	14.46
S11	0.20	89.53	10.27
S12	0.83	83.43	15.74
S13	0.20	88.25	11.55
Average	0.59	85.96	13.45
±STD	0.34	2.85	2.80

Conclusion

In this investigation, petroleum hydrocarbons known as *n*-alkanes were found in samples taken from the Selangor River. Additionally, using various molecular tools, the content and distribution of these hydrocarbons were examined and their sources were identified. The results of the investigation show that the n-alkane levels are nearly equal at all sites. The L/H *n*-alkanes ratio values were frequently greater than 1 in the sediment samples. The investigation's findings also revealed that the majority of the silt included *n*-alkanes with an even number of carbon atoms, which can be attributed to microbial activity. The results of this investigation demonstrated that *n*-alkanes in sediments originated from a range of sources. Biogenic materials, such as leftover algae, aquatic life, terrestrial plants, and human activity, are potential sources of *n*-alkanes. The prevalence of HMW n-alkane with odd numbers of carbon atoms in certain of the sediment sample locations can be attributed to input of n-alkane from terrestrial high plants. In conclusion, the data suggested that the water quality of the Selangor River has declined as a result of anthropogenic activities in the state and rapid population growth. The concentrations of the investigated hydrocarbons varied from one sampling site to another and from one time period to another. As a result, ongoing assessment of the degree of the impact of hydrocarbon pollution on human or ecosystem health can show the level of pollutants brought about by anthropogenic activities. As a result, more research into hydrocarbons and other anthropogenic contaminants is essential for minimizing health hazards in Peninsular Malaysia.

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Declarations

Competing interests The authors declare no competing interests.

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