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Assessment of Sewage Molecular Markers: Linear Alkylbenzenes in Sediments of an Industrialized Region in Peninsular Malaysia

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Abstract: In this study, the use of linear alkylbenzenes (LABs) was employed to pinpoint the sources of human activity that cause detrimental impacts on the coastal environment and river ecosystems. LABs were detected using GC–MS in sediment samples assembled from Kim Kim River (KKR) and the Port Dickson coast (PDC). To assess the significance of variations in the distribution and concentrations of LABs across the sampling sites, this study utilized several statistical techniques such as post hoc tests, LSD techniques, analysis of variance (ANOVA), and the Pearson correlation coefficient using a significance level of $p < 0.05$. The degradation levels of LABs and wastewater treatment were assessed in the study using internal congeners (I/E), homologs of C^{13} and C^{12}, and long-to-short-chain (L/S) ratios. The results revealed that the LAB concentrations varied between 88.3 and 112 ng/g dw in KKR and 119 to 256 ng/g dw in the PDC. Most of the surveyed areas exhibited a substantial count of C^{13}–LABs homologs that displayed a significant difference ($p < 0.05$). The I/E ratios ranged from 1.7 to 2.0 in KKR and from 2.0 to 4.1 in the PDC, suggesting that the effluents originated from sources associated with the physical phase and biological phase in wastewater treatment systems (WWTSs). The results revealed that the degradation of LABs varied between 34% and 38% in KKR and between 40% and 64% in the PDC. This study underscores the importance of ongoing improvements to WWTSs and emphasizes the potential of LABs as indicators for monitoring wastewater contamination.

Keywords: linear alkylbenzenes; sediment; internally over exterior; pollutants contained in wastewater; degradation; molecular marker

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1. Introduction

Anthropogenic pollution stemming from metropolitan areas is a significant contributor to the degradation of riverine and coastal ecosystems [1–4]. The release of substantial amounts of industrial and household wastewater in these regions significantly affects the biodiversity of the ecosystems [5–8]. Therefore, regular monitoring of aquatic ecosystems and water sources is crucial for maintaining the health and integrity of these environments [9–14]. It provides valuable data for decision making, pollution control, and the preservation of biodiversity, ensuring sustainable management and protection of our precious water resources [15–18]. Obtaining such data is crucial for achieving efficient environmental security and economic management. With this information, strategies and implemented measures can be developed that mitigate the harmful impacts of anthropogenic pollution on these delicate ecosystems, helping to preserve them for future generations [8,19–22].

Linear alkylbenzenes (LABs) can serve as indicators of industrial and domestic pollution. Linear alkylbenzene sulfonates (LASs), which constitute the primary ingredients of detergents, are composed of alkyl chains ranging from C10 to C14 and are referred to as LABs [23]. In the 1960s, the utilization of branched alkylbenzene constituents was replaced by LABs, which were preferred for their cost-effectiveness and biodegradability. However, ref. [24] noted that improper sulfonation has led to the extensive and prevalent discharge of LABs into the aquatic ecosystem through untreated wastewater effluent. In contrast, ref. [25] indicated that LABs exhibit a significant attraction toward industrial effluents, which renders them valuable as indicators of pollution from human-made sources, including both residential and industrial, in aquatic settings.

LAB isomers, which have varied phenyl substitutions and internal/external isomers alongside the straight alkyl chain, serve a purpose in gauging degradation in sediment and suspended particulate matter, as described by [26]. Typically, wastewater treatment systems (WWTSs) contain a series of physical (primary treatment) and biological (secondary treatment) processes designed to remove contaminants from wastewater before it is discharged back into the environment or reused for other purposes [27]. The specific components of a wastewater treatment system can vary depending on the type of contaminants present in the wastewater and the desired level of treatment. According to [28], the identification of industrial and domestic wastewater types discharged into marine ecosystems, including physical (primary) and biological (secondary) systems, as well as the estimation of pollutant residence time, can be accomplished through the utilization of LAB isomers.

Due to the high population in coastal areas of Malaysia, there has been a significant increase in industrial and domestic wastewater contamination. Studies in Asian countries demonstrated a positive correlation between sewage contamination and illnesses [29]. Therefore, it is necessary to investigate the impact of marine sediments and their fate on the possible pollution caused by wastewater to improve water quality and reduce the risk of illness [30,31]. Based on three local media that were recently published, there is water contamination in Simpang Renggam in Sungai Benut, which mostly affects 75,000 residents in Johor state [32]. Thus, the aquatic environment around West and South Malaysia should be frequently monitored to identify the sources of contamination, whether industrial or home wastewater, in order to protect environmental and human health.

Regular monitoring of wastewater sources in West and South Malaysia is crucial for protecting the environment and human health. Although the anthropogenic impacts in KKR and the PDC were investigated, it is essential to extend the research to other regions. This study employed LABs as anthropogenic pollution to evaluate the effects of wastewater sources in the selected areas. This study measured the concentration, distribution, and degradation levels of LABs in the study regions, and investigated the distribution of LAB isomers to enhance the effectiveness of existing wastewater treatment systems (WWTSs).

2. Methods and Materials

2.1. Experimental Design

The research sites that were studied were situated in the southwest region of Malaysia, specifically in the states of Johor and Sembilan, as shown in Figure S1 in the Supplementary Information (SI). Seven sediment samples were taken from sites chosen for their proximity to agricultural and industrial activity, such as Kim Kim River in Johor Bahru State, as well as urban and tourism activity, such as Port Dickson Beach in Sembilan State. Details and a visual representation of the sampling locations used in the study are provided in Table S1 in the SI file. To examine the presence of LABs in these areas and to assess the level of anthropogenic contamination, surface sediments were collected from the top 4 cm of KKR and the PDC using an Ekman dredge sampler. The collected sediment was then put into a stainless steel container that had been cleaned, and the top 4 cm of the sediment cake was sliced, immediately kept in a double-clean Ziploc bag, labeled, and then put on ice at 4 °C in a cooler box for transport to the lab, where it was stored at -20 °C upon arrival. The samples were then put through a freeze-drying procedure to remove any remaining moisture and get them ready for more analysis.

2.2. Chemical Analysis

Two columns were used to extract LABs. The initial one was used for purification to remove polar compounds, and the second was used for fractionation to obtain the desired organic extracts, following the established validation protocols described elsewhere by [33]. To recover dried sediments, 10 g of sediments were mixed with 250 mL dichloromethane (DCM) in a cellulose thimble. This mixture was left for 8–10 h, as per the protocol in [34]. The surrogate standards (SSs), which are similar to the target analytical compounds used in the analytical method for the purpose of identification and could be recovered from the sample matrix with reasonable efficiency, were preloaded in each sample to ensure the precise recovery of the target LABs. The SSs contain 50 µL of 1-C*n* LABs, where 1- denotes each LAB homolog's initial isomer and *n* denotes its carbon number (8–14). Activated copper was added to remove any sulfur that could interfere with the GC chromatograms. The extract was concentrated using a rotary evaporator and then placed on a chromatography column of 5% H_2O-deactivated silica gel (60–200 mesh size, Sigma Chemical, St. Louis, MO, USA), with a diameter of 0.9 cm and height of 9 cm. Hydrocarbons were eluted from the column with a pure hexane/DCM mixture (3:1, v/v) in 20 mL aliquots and then reduced to 1–2 mL. LAB fractions were collected from a fully activated silica gel column with a diameter of 0.47 cm and height of 18 cm using 4 mL of hexane in the second stage. The reduced LAB fractions were transferred to a 2 mL amber vial and dried using a moderate stream of nitrogen. Before performing the GC–MS analysis on the LABs fraction, an internal standard (IS) of biphenyl–d10 with a mass-to-charge ratio (m/z) of 164 was injected.

To detect compounds of LABs, a gas chromatograph from the Agilent Technologies (Santa Clara, CA, USA) 7890A series was utilized, along with a C5975 MSD split/splitless injector. The experiment utilized a 30-meter-long fused silica capillary column with an inner diameter of 0.25 millimeters and a DB–5MS capillary column coated with a 0.25 µm film thickness. Helium was chosen as the carrier gas and was kept at a steady pressure of 60 kg cm^{-2} throughout the experiment.

Further, to obtain the mass spectrum data, the LABs chromatographs were monitored at m/z = 91, 92, and 105 using a selective ion monitoring (SIM) mode. The method utilized for this data operated with specified parameters, such the signal-to-noise ratio (S/N), peak detection limits, and a 5-point calibration curve. LAB peaks in the samples were obtained by comparing the retention times of each target compound to the retention times of the LAB standards run on the same day, which were then confirmed using the m/z value (91, 92, and 105). An external source was employed to carry out the ionization process at a temperature of 200 °C. The GC–MS operating conditions were configured with an electron energy of 70 eV and an electron multiplier voltage of approximately 1250 eV. After injecting the sample using the splitless mode, the injection port temperature was maintained at

300 °C, and a one-minute purge ensued. The temperature T of the column was maintained at 70 °C for two minutes. It was then raised to 150 °C at a rate of 30 °C per minute and subsequently increased at a rate of 4 °C per minute until it reached 310 °C. This final temperature was maintained for 50 min.

2.3. Quality Control and Assurance

LABs are used to determine the degree of degradation due to the properties of external and internal isomers. External isomers degrade faster than internal isomers due to the substitution position of the phenyl group on the straight alkyl chain. Furthermore, LABs are persistent in aquatic environments for extended periods. The I/E ratio can be used to identify the types of wastewater treatment. For instance, raw sewage has a lower I/E ratio, ranging from 0.7 to 0.8. Primary treatment I/E ratios vary between 0.5 and 0.9, while secondary treatment has an I/E ratio greater than 2 [35]. The LABs surrogates (1–C*n*) were successfully recovered in the 60–120% range with minimal loss of the target chemicals during the analytical procedures. The recovery rates for all sediment samples tested were between 87 and 98%.

To ensure that there was no contamination, a blank sample was included in every batch of four samples, which contained all the substances and standards found in typical samples. Each day, the sediment samples were spiked with freshly made surrogate, internal, and native standards at predetermined amounts. The target LAB congeners were located using GC–MS in selected ion monitoring mode at $m/z = 91$, 92, and 105. The quantification of the LAB chemicals was done using a 5-point calibration curve with a LABs standard mixture, with concentrations ranging from 0.25 to 5.0 ppm spiked into the target samples. The limits of quantitation (LOQ) and detection (LOD) were determined based on the lowest concentration levels of each calibration curve divided by the mean sample weight, which were found to be 0.02–0.1 ng/g and 0.1–2 ng/g, respectively, following the method described by [36].

2.4. Statistical Analysis

The statistical analysis process was undertaken using the IBM®–SPSS 25 program. Various techniques, including analysis of variance (ANOVA), post hoc tests, and LSD, were utilized to showcase the significance of fluctuations in the LABs concentrations and distributions across sample sites at a significance level of $p < 0.05$. To evaluate the relationship between sampling stations, the Pearson correlation coefficient was utilized. Additionally, principal component analysis (PCA) was conducted to decrease the number of initial variables, such as the concentration of LABs, and identify a smaller set of underlying factors. The extraction process retained only those elements that had eigenvalues greater than one, which were then subjected to varimax rotation. Standardized regression coefficients were utilized to assess the potential contribution of various sources of LABs.

2.5. Evaluation of Total Organic Carbon (TOC)

The TOC in sediment samples was detected using LECO CR–412 analysis. The samples were dried at 60 °C overnight and then pulverized using a mortar and pestle. In order to remove any inorganic carbon, sediment samples weighing 1–2 g were fully moistened with 2 mL of 1 M HCl, which eliminates carbonates. The sediment samples were then dried once again at 100 °C for 10 h to remove the HCl. Finally, the TOC % was determined using a LECO CR–412 Carbon Analyzer, which had a furnace temperature of 1350 °C and an O_2 boost period of 1 min. The procedure described in [36] was followed.

3. Results and Discussion

3.1. Differences in the Distribution of LABs Concentrations

The LABs formula, depicted in Figure S2, is denoted as "n–Cm", with "n" indicating the position of the benzene ring on the linear alkyl chain and "m" indicating the number of alkyl carbons. The sediment samples that were analyzed consisted of LABs with carbon chain lengths between C10 and C14. Concentrations of LABs in KKR and the PDC were found to be between 88.3 and 112 and between 119 and 256 ng/g dw, respectively. The huge variation, especially among the samples from the PDC, was attributed to anthropogenic activities near the sampling stations, such as urbanization growth, small industries, tourism, and shipping.

According to the study's findings, the significant Pearson correlation between the LAB concentrations in the investigated areas ($r = 0.88$, $p < 0.05$) was most likely caused by the fact that LAB concentrations were higher in areas with a high population density, industrial, agricultural, and tourism activities, as these areas are more likely to have a high volume of wastewater and detergent discharge. Overall, the strong correlation between LAB concentrations and pollution indicates that LABs could serve as a useful measure of both residential and industrial contamination. Table S2 in the SI file shows the details of this correlation. The range of LAB levels was between 88.3 and 256 ng/g dw, with significantly higher concentrations observed at the PDC line, as illustrated in Figure 1. Additionally, the statistical analysis at $p < 0.05$ revealed a significant difference in the LABs concentrations between the various study locations (refer to Table 1).

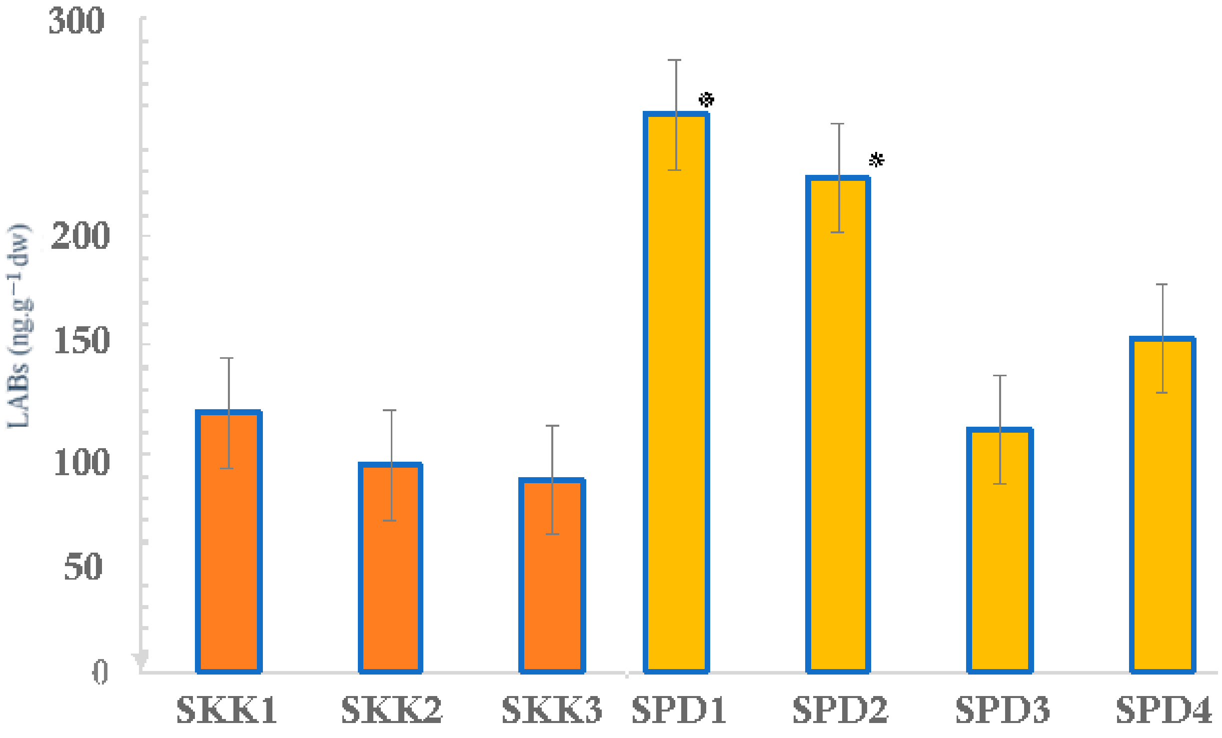

Figure 1. Concentrations of LABs in the Kim Kim River and Port Dickson samples. Standard error bars are shown. (*) indicates the highly significant differences ($p < 0.05$).

The distribution of LABs in riverine environments may be linked to treated effluent runoff and the dilution of organic matter, as indicated by their content. This can affect the distribution of riverine ecosystems, as observed by [37,38]. On the other hand, according to studies by [39,40], the occurrence of LABs in coastal areas is thought to result from high levels of industrialization and urbanization, as well as the lack of piped sewers. In contrast with previous studies by [41,42], which found lower concentrations of LABs at their research sites compared with Port Klang, the Penang Estuary, Malacca, and the Anzali Wetland, refs. [43,44] reported similar lower levels at the respective locations. Nonetheless, the present study identified relatively high quantities of LABs, comparable with those found in Southern Brazil and the Pearl River. The higher levels of LABs detected in this study compared with some other regions worldwide and Malaysia could be attributed to a combination of specific local factors and differences in pollution sources, such as significant industrial, tourism, and urban activities that release LABs into the environment. Port

Dickson is known to be one of Malaysia's famous recreational beaches, with numerous hotels and resorts that offer a wide range of accommodations, as well as the growth in urbanization, small industries, tourism, and shipping [45]. The distribution of LAB isomers and homologs among this trend suggested that their presence was uniformly distributed at the KKR and PDC sites, which implies that the geographic location of the sampling locations may have influenced the distribution of LABs. Additionally, the LABs detected in this study were found to be moderate to low compared with other regions worldwide and Malaysia, suggesting that they may serve as an indicator of wastewater pollution.

Table 1. LABs concentrations (ng/g dw) and relative compound ratios in Kim Kim River and the Port Dickson coast.

Compound	SKK1 [b]	SKK2	SKK3	SPD1	SPD2	SPD3	SPD4
C_{10}-LABs ($ng{\cdot}g^{-1}$ dw) [a]	9.4	7.7	6.8	17.2	15.0	4.2	6.2
C_{11}-LABs ($ng{\cdot}g^{-1}$ dw)	16.3	13.0	12.0	66.7	57.0	19.0	30.7
C_{12}-LABs ($ng{\cdot}g^{-1}$ dw)	25.6	20.7	18.6	68.6	61.0	24.0	38.0
C_{13}-LABs ($ng{\cdot}g^{-1}$ dw)	41.0	32.6	29.5	86.0	78.4	51.2	63.6
C_{14}-LABs ($ng{\cdot}g^{-1}$ dw)	26.8	21.1	21.3	17.4	15.6	13.2	14.8
LABs ($ng{\cdot}g^{-1}$ dw)	119	95.2	88.3	256	227	112	153
LC-LABs ($ng{\cdot}g^{-1}$ dw) [c]	67.8	53.8	51.0	103	94.0	64.5	78.4
SC-LABs ($ng{\cdot}g^{-1}$ dw) [d]	25.8	20.7	19.0	84.0	71.8	23.2	37.0
I/E [e]	2.0	1.8	1.7	4.1	3.7	2.0	2.6
L/S [g]	2.4	2.5	2.6	1.6	1.6	1.7	1.6
C_{13}/C_{12} [h]	5.1	5.2	5.2	9.7	9.5	13.7	14.3
LAB degradation (%) [f]	38	35	34	64	61	40	48
TOC (%) [i]	1.2	1.0	8.1	2.0	2.3	1.5	1.9
TOC (mg/g)	11.5	9.0	81	19.7	22.7	14.7	18.9

Notes: [a] C10-LAB: sum of the 26LAB congeners ranging from 5-C10 to 2-C10. [b] SKK1–3 and SPD1–4: the first letter indicates the station; the second and third letters represent the first letters of location name; and the numbers 1, 2, 3, and 4 indicate the first, second, third, and fourth station for each respective location. [c] LC-LABs: sum of LABs ranging from 6-C^{13} to 2-C^{14}. [d] SC-LABs: sum of LABs ranging from 5-C^{10} to 2-C11. [e] I/E (C^{12}–LABs): ratio of (6-C^{12}LAB + 5-C^{12}LAB) relative to (4-C^{12}LAB + 3-C^{12}LAB + 2-C^{12}LAB). [g] L/S: ratio of (5-C^{13}LAB + 5-C^{12}LAB) relative to (5-C^{11}LAB + 5-C^{10}LAB). [h] C^{13}/C^{12}: ratio of (6-, 5-, 4-, 3-, and 2-C^{13}/(6-, 5-, 4-, 3-, and 2-C^{12}LAB). [f] LAB degradation (%): LAB deg = 81 × log (I/E ratio) +15 ($r^2 = 0.96$). [i] TOC (%): total organic carbon.

Table 2 indicates that there is a notable disparity in the allocation of LAB homologs among the different sampling locations, with a statistical significance level of $p < 0.05$. The geographical features of different sampling locations, such as proximity to coastlines, rivers, or urban centers, as well as industrial discharges, urban runoff, or agricultural activities and inadequate treatment, could influence the input sources of LAB homologs and their subsequent distribution [46]. The LABs in KKR contained a higher proportion of homologs and isomers with C^{13}, followed by C^{12} and C^{14} (Figure 2), indicating a substantial difference in composition compared with other locations. Conversely, the LABs containing C^{10} and C^{11} were present at low levels, suggesting selective degradation of these compounds during industrial effluent discharge. LC–LABs (long-chain linear alkylbenzenes), such as C^{13} and C^{14}, were found to be the primary component of LABs concentration vs. SC–LABs (short-chain linear alkylbenzenes), like C^{10} and C^{11}, in the river. The chemical test indicated that the concentrations of isomers, such as 6-, 5-C^{13}, and 6-C^{12} were higher than those of other isomers, suggesting that LC–LABs had been transported over a long distance. At the SKK1 station, refs. [47,48] observed that a high concentration of C^{13} homologs was found, suggesting anaerobic degradation of these compounds. Surprisingly, this study revealed a higher concentration of LABs than reported by [49], with numerous LABs found in the mixture of molecules that are isomeric and homologous, particularly LC–LABs, such as C^{13}–LABs.

Table 2. Total LABs concentration in sediments among different locations of Kim Kim River and Port Dickson, Southern Malaysia: (**a**) ANOVA and (**b**) post hoc tests.

(a) ANOVA					
	Sum of Squares	**df**	**Mean Square**	**F**	**Sig. ***
Between groups	8410	4	210	7.3	0.000
Within groups	8680	30	289		
Total	17,100	34			

(b) Post Hoc Tests

Dependent Variable: LAB Concentration

LSD

(I) Location	**(J) Location**	**Mean Difference (I-J)**	**Std. Error**	**Sig.**	**95% Confidence Interval**	
					Lower Bound	**Upper Bound**
1.00	2.00	−21.2 *	9.1	0.03	−39.7	−2.6
	3.00	−27.2 *	9.1	0.01	−45.7	−8.6
	4.00	−45.1 *	9.1	0.00	−63.7	−26.6
	5.00	−9.1	9.1	0.32	−27.7	9.5
2.00	1.00	21.2 *	9.1	0.03	2.6	39.7
	3.00	−6.0	9.1	0.52	−24.6	12.6
	4.00	−24.0 *	9.1	0.01	−42.5	−5.3
	5.00	12.1	9.1	0.20	−6.5	30.6
3.00	1.00	27.2 *	9.1	0.01	8.6	45.7
	2.00	6.0	9.1	0.52	−12.6	24.6
	4.00	−18.0	9.1	0.06	−36.5	0.6
	5.00	18.0	9.1	0.06	−0.53	36.6
4.00	1.00	45.1 *	9.1	0.00	26.6	63.7
	2.00	24.0 *	9.1	0.01	5.4	42.5
	3.00	18.0	9.1	0.06	−0.6	36.5
	5.00	36.0 *	9.1	0.00	17.4	54.6
5.00	1.00	9.1	9.1	0.32	−9.5	27.7
	2.00	−12.1	9.1	0.20	−30.6	6.5
	3.00	−18.0	9.1	0.06	−36.6	0.53
	4.00	−36.0 *	9.1	0.00	−54.6	−17.4

Notes: * The mean difference was significant at the 0.05 level.

The PDC sediments contained the highest concentration of C^{13}–LABs homologs while having the lowest levels of C^{10} homologs when compared with all other sites in the region. The distribution of LABs was dominated by LC–LABs, like C^{13} and C^{14}, with higher concentrations than SC–LABs, like C^{10} and C^{11}. The order of homologs by concentration was C^{13}, C^{12}, C^{14}, C^{10}, and C^{11}. According to [50], the variation in composition within this range is probably caused by the growing hydrophobic nature of LABs with longer chains. In contrast with the detergents analyzed by [41], the sediments in this region had a reduced amount of C10 homologs in their distribution. Figure 3 illustrates the contrast between the LC–LABs and SC–LABs in terms of the areas studied, allowing for a comparison between them. Therefore, LC homologs were discovered to be more plentiful than SC homologs in the first and second sites. This contrast may be attributed to the greater decline in SC homologs, particularly C^{10} and C^{11}, in the sediments, as documented by [51,52]. LC–LABs, being longer-chain compounds, are generally less volatile and more hydrophobic compared with SC–LABs, and this can result in differences in their environmental fate and transport. LC–LABs may have a greater tendency to adsorb onto particles and sediments due to their higher molecular weight and hydrophobicity. This enhances their sorption and partitioning into sediment matrices, making them more likely to be retained in the sediments over time. LC–LABs also might undergo slower degradation or transformation processes due to their larger size and more complex structure [53,54].

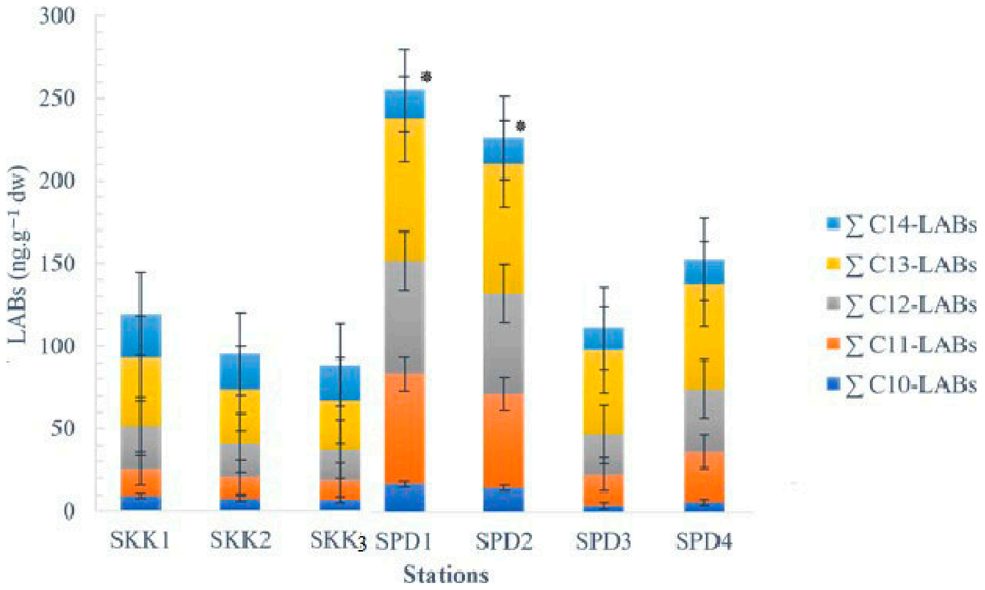

Figure 2. Compositional profiles of LABs in the Kim Kim River and Port Dickson coast sediments. (*) indicates the highly significant differences ($p < 0.05$).

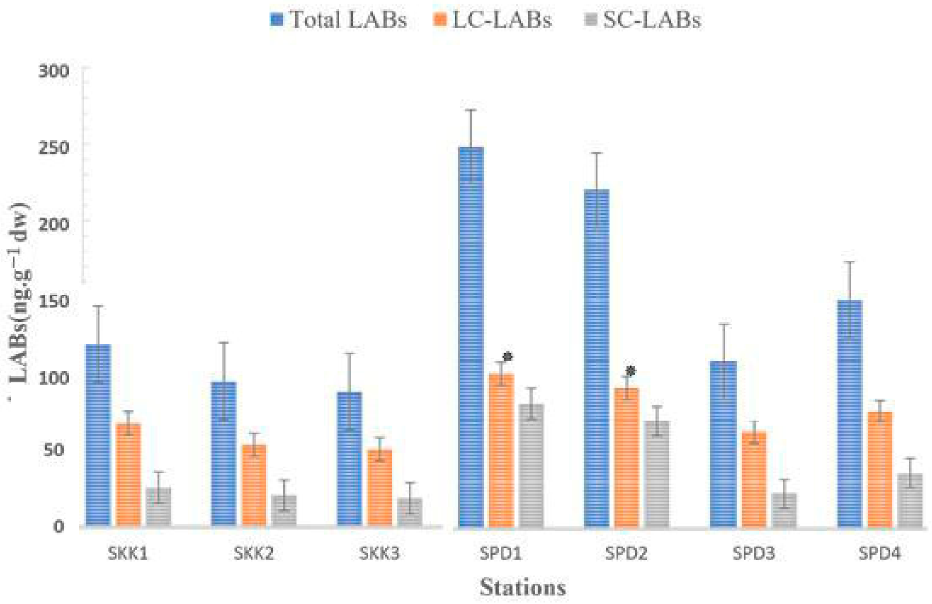

Figure 3. Concentrations of short-chain (SC-LABs), long-chain (LC-LABs), and total linear alkylbenzenes (LABs) in sediments from the Kim Kim River and Port Dickson samples. (*) indicates the highly significant differences ($p < 0.05$).

3.2. Assessing the Efficiency of LABs Removal from Wastewaters

According to [24], the LABs in KKR mainly originated from physical and biological treatment effluents due to the high levels of WWTSs present in the study areas. Figure 4 in their study provides evidence to support this claim. The I/E ratios in KKR varied between SKK2 and SKK1, with an average of 1.8, indicating that primarily treated effluents were released into the river water. This finding is consistent with the study conducted by [55]. On average, the L/S ratio fell between 2.4 and 2.6, with detergents showing a ratio higher than 1.8, indicating that LABs were effectively biodegraded [56]. Furthermore, KKR and the PDC had an average C13/C12 ratio ranging from 5.1 to 5.2, with an average of 5.1, surpassing the ratio of 1.7 recorded in coastal sediments.

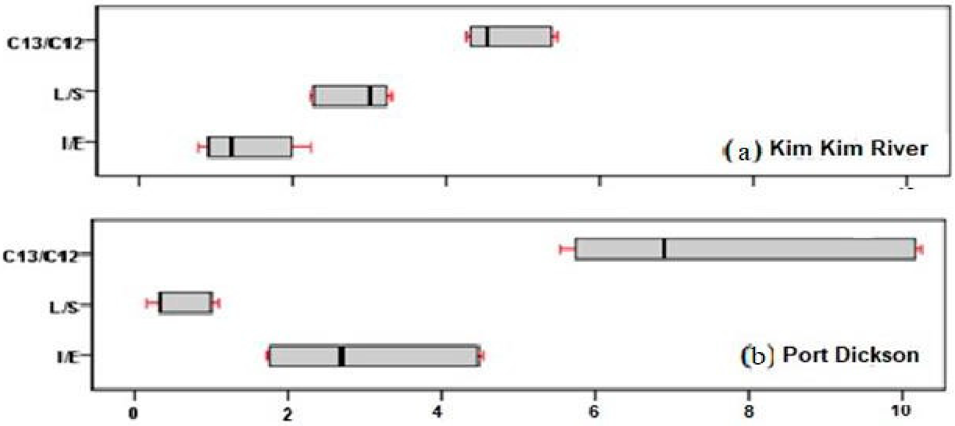

Figure 4. Values of the I/E, L/S, and C^{13}/C^{12} ratios in the samples of Kim Kim River and Port Dickson.

On the other hand, the PDC had an I/E ratio ranging from 2.0 to 4.1, which indicated the discharge of biological treatment effluents in the region. These ratios were considerably higher than those reported by [34] in Selangor's river (0.2–1.0), indicating a substantial discharge of pre-treated effluents in the PDC. Additionally, the degradation of LABs was observed to occur under aerobic conditions. In order to assess the biodegradation of LABs in the studied areas, L/S and C^{13}/C^{12} ratios were used. The presence of LABs supports the idea that untreated effluents were discharged into the PDC, as previously mentioned. According to [57,58], over 80 sewage pipelines were identified as sources of untreated wastewater and effluent discharge from residential areas and hotels, resulting in a deterioration of coastal quality and protection.

The degradation rates of LABs showed variation across different locations, with lower rates found in KKR (SKK3, 34%) and higher rates in the PDC (SPD1, 64%). The results suggest that LABs degrade faster along the PDC shore than in KKR. Over the past few years, waste discharge from ferries and boats, which are used for recreational and fishing purposes, directly into KKR has led to an increase in LABs in the river sediments due to the disposal of detergent waste and washing of boats. These activities have also significantly impacted molecular indices. Untreated wastewater discharge from physical and biological treatments in WWTSs is a significant cause of LAB substances in Malaysia. The results of studies by [59,60] indicate that the KKR region has higher concentrations of industrial effluents, and it discharges LABs from both domestic and industrial wastewater sources. These findings suggest that there could be increased transportation of LABs from their sources to subsequent stages.

3.3. Investigation of TOC

Due to LABs being hydrophobic compounds, they tend to strongly adhere to organic materials in aquatic environments. Consequently, the concentration of LABs found in sediments is closely correlated with the quantity of the TOC available [61]. Thus, a weak correlation ($R_2 = 0.42$) was discovered between the LABs and TOC in sediment samples from

KKR. However, linear regression analysis showed that the TOC did not have a significant impact on the distribution of LABs. The primary factor determining the distribution of LABs in the area was found to be the intensity of their release from various wastewater sources. Previous studies conducted by [34] in Selangor, Peninsular Malaysia, and by [62] in Perak Rivers reported modest correlations between LABs and the TOC, with different R_2 values (0.008 and 0.17, respectively). Nevertheless, the PDC revealed a correlation (R_2 = 0.64) between the concentration of LABs and the TOC, indicating that the TOC could be a significant factor in determining the spatial distribution of LABs from urban and industrial sources. This discovery aligns with findings from Dongjiang River (R_2 = 0.82), which suggests that industrial and domestic WWTSs are essential sources of organic matter.

3.4. Source Apportionment

During the investigation, significant fluctuations were observed in the I/E ratio, with readings below 1 at many sites, suggesting the discharge of untreated wastewater into the aquatic environment. In order to identify potential sources of LABs in KKR and the PDC shoreline, according to the Kaiser criterion, the data variability at the measurement points can be described by two components. The performed calculations showed that PC1 explains 74.40% and PC2 explains 18.27% of the variability. C10–C14-LABs, LC-LABs, SC-LABs, I/E, L/S, and latitude had the greatest influence on the PC1 component, with the factor loadings ranging from −0.74 to 0.99. However, these variables resulted in a decrease in the PC1 value (Figure 5 and Table S3 in the SI file). On the other hand, the PC2 component was mainly influenced by C13/C12 (−0.815), with a smaller influence from C10-LABs and C14-LABs (0.671–0.711), which led to an increase in the component. The strong correlation between C12–C11–SC-Labs-LC-Labs_Degr was confirmed by the closeness between the variables in Figure 5. Additionally, an increase in longitude and LS led to a decrease in C13/C12. Finally, it was established that there was no correlation between C10 and longitude or latitude.

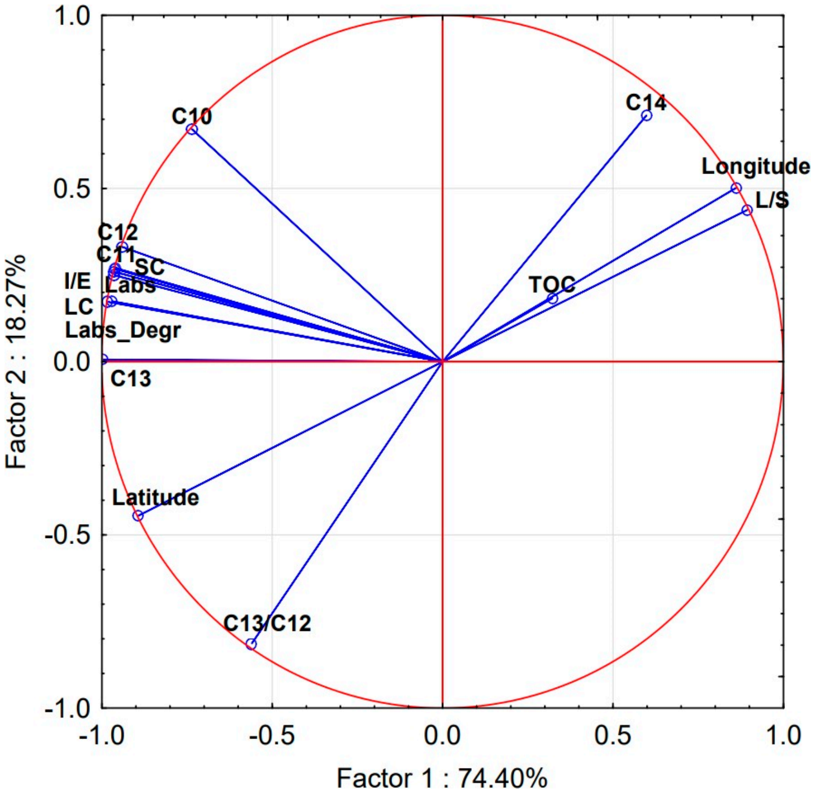

Figure 5. Relationships between factor 1 and factor 2 for loading variables in the sediment samples of Kim Kim River and Port Dickson coast.

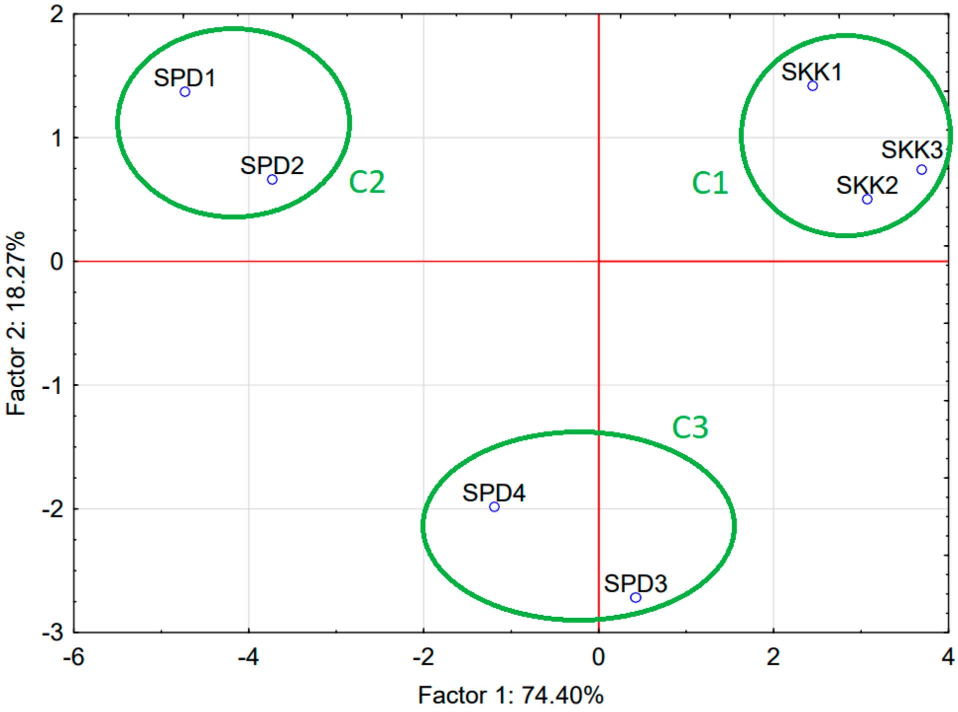

Figure 6. Projection of measurement points on the plane of factors 1 and 2 for loading variables in the sediment samples of Kim Kim River and the Port Dickson coast.

4. Conclusions

The concentration of sedimentary LABs varied greatly among the locations studied, ranging from 88 to 256 ng/g dw. Compared with KKR, the PDC had higher LABs concentrations with more LC homologs than SC homologs. Physical treatment effluents were found to be the essential source of LABs in KKR, while biological treatment effluents dominated the PDC, as indicated by the I/E ratios. The high LABs concentrations in the sediment indicate that the current WWTSs may not be capable of handling the rising population in the area. This research highlights the ongoing problem of untreated wastewater sources, i.e., domestic sewage and industrial influent, which could pose increasing risks as the population expands. According to future projections, the discharge of industrial and urban effluents into rivers and beaches in Peninsular Malaysia is expected to keep rising. This underscores the urgency of enhancing the WWTSs. The findings of this study shed light on the level of pollution in the studied regions, as well as its distribution and sources of organic pollutants (LABs). Public knowledge of the pollution issues in the studied regions can be improved, support for pollution control measures, and environmentally responsible behavior can be encouraged by sharing the findings of environmental monitoring studies. This information can be used to inform decisions made by the authorities about the implementation or modification of pollution control regulations. To decrease sewage contamination in the near future, more effective STPs are required in neighboring areas, together with ongoing monitoring of the marine environment to prevent potential contamination from sewage pollution.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15183301/s1.

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