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Lateral Distribution, Environmental Occurrence and Assessment of Organic Pollutants in Surface Sediments of the West and South Peninsular Malaysia

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Abstract Linear alkylbenzenes (LABs) are molecular chemical markers that are used to identify anthropogenic inputs in rivers and coastal ecosystems. Surface sediment samples had collected from the west and south of Peninsular Malaysia to determine the LAB content and distribution. The samples were analyzed, and their sources were determined using gas chromatography-mass spectrometry (GC-MS). The statistical significance of the variations among sampling stations was examined using the analysis of variance (ANOVA) and Pearson correlation coefficient at $p < 0.01$. The degrees of LAB degradation and the effectiveness of sewage treatment were assessed using long to short chains L/S, homologs C_{13}/C_{12}, and internal to external (I/E) indices. The statistical analysis revealed that the concentration of LABs in the areas under investigation ranged from 67.4 to 255.8 ng g^{-1} dw. LAB homologs were significantly different at $p < 0.01$, and the majority of sampling sites had a large number of C_{13}-LAB homologs. It was clear that treated effluents from main and secondary sources were released into the study areas since the computed LAB ratios (I/E) ranged from 1.6 to 4.1. In addition, LAB deterioration ranged between 33 and 64% in the regions studied. It is concluded that the wastewater treatment system needs to be upgraded, and there is a strong need to use LAB molecular markers to trace anthropogenic sewage contamination.

Keywords Industrial · Organic marker · I/E · Contamination · Sediment

1 Introduction

Riverine and coastline environments situated near urban locations receive frequently heavy quantities of anthropogenic inputs (Halpern et al., 2008; Hagemann et al., 2018), However, release high loads of untreated industrial effluents into these areas lead to change the biodiversity in coastal and marine environments (Zhang et al., 2012; Melo et al., 2019). The monitoring of these ecosystems is important to assess the occurrence of possible hazardous contaminants as well as related adverse influences to marine ecosystem, hence presenting the necessary data for the possible environmental protection (Chaler et al., 2004;).

Domestic sewage pollution can be measured using chemical compounds such as linear alkylbenzenes (LABs). The main components used to make linear alkylbenzene sulfonates type detergents (LASs) are LABs (Ricking et al., 2003). In the 1960s, branched alkylbenzene components were totally switched to

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LABs for detergent manufacturing due to their excellent biodegradability and low cost (Sherblom et al., 1992). As a result of insufficient sulfonation, LABs are widely dispersed and abundant in the aquatic ecosystem from non-treated residential and industrial wastes (Oller et al., 2011; Alkhadher et al., 2020). Considering their enduring and high affinities for wastewater effluents, LABs are used as indicators of anthropogenic input in the marine environment that are coming from residential and industrial sources (Cabral et al., 2020; Alkhadher et al., 2020).

Because they have different phenyl replacements to the alkyl chain and are organized externally and inside, LABs are also employed to show the degree of degradation in the sediments (Alkhadher et al., 2015; Huang et al. 2021). Furthermore, the types of industrial and residential wastewater discharged into an aquatic ecosystem, such as primary and secondary effluents, had determined using LAB isomers (Takada et al., 1990; Tsutsumi et al., 2002).

Due to the country's dense population and the relationship between sewage pollution and illnesses in East Asian nations, Malaysia's coastal areas have demonstrated significant levels of residential wastes (Isobe et al., 2002; Sakai et al., 2017; Thomes et al., 2019). As a result, it is crucial to regularly assess the anthropogenic LAB inputs and their impacts on coastal locations in order to enhance water quality and reduce the risk of disease (Wang et al., 2001; Alkhadher et al., 2020).

Rivers and coasts of west and south Malaysia are adjacent to populous as well as developed areas that leads to land-based pollutants include municipal sewage, agricultural runoff, and industrial waste into several rivers and coastal ecosystem (Alkhadher et al., 2016; Masood et al., 2021). Because of this, the rivers and beaches in this area may be impacted by various anthropogenic wastes. Thus, the sources of industrial and domestic wastewater pollution of west as well as south Malaysia should monitor continuously for an environment protection as well as people health. Nevertheless, the investigation of the recent anthropogenic impacts is conducted in the west as well as south Malaysia, which they have industrialized and populous characteristics in some locations such as the Port Dickson, Muar river, Johor Bahru coast, as well as Kim Kim river while they are less industrialized or unpopulated in the other locations such as Pulau Merambong. The current study goals to use LAB as chemical tracer to evaluate an industrial and residential wastewater contamination in the selected stations. Measuring of concentration, distribution, as well as LAB degradation level carried out in abovementioned areas. Additionally, the dispersion of LAB isomers was carried out in order to identify the performance of existing sewage treatment plants (STPs).

2 Methods and Materials

2.1 Sampling

The researched stations are located in the Sembilan and Johor states, including rivers, estuaries, islands, and coasts alongside of west as well as south Peninsular Malaysia (Fig. 1). The description of the study locations of the Port Dickson coast with station code (SPD1, SPD2, SPD3, SPD3, SPD4), Muar river with station code (SMu1, SMu2, SMu3), Johor Bahru coast with station code (SJB1, SJB2, SJB3, SJB4, SJB5), Pulau Merambong with station code (SMe1, SMe2, SMe3), and the Kim Kim river with station code (SKK1, SKK2, SKK3) are illustrated in Table 1. The studied areas have chosen based on the occurrence of less and high anthropogenic activities alongside south and west peninsular Malaysia. The collection of the surface sediments conducted to evaluate the most recent anthropogenic LAB imports in the aforementioned regions. To analyze the current LAB inflow, the top 4 cm of each surface sediment had collected using an Ekman dredge sampler (Ibrahim, 1988). All samples were then put in pre-solvent washed stainless steel containers before being brought to the lab and kept in a freezer at 20 °C. In order to prepare the sediments for further chemical analysis, a freeze-dryer is employed to remove the moisture.

2.2 Chemical Procedures

Two columns had used for purification and fractionation the sediment extracts in order to analyze the LABs (Zakaria et al. 2002). In particular, within a Soxhlet device, 10 g of dried sediment was placed in a cellulose thimble, followed by 250 mL of dichloromethane (DCM) for 10 h (Masood et al. 2015). Prior to extraction, 50 L of "1-Cn" LABs had added to each sample as surrogate standards (SS) for the recovery correction of target LABs.

Fig. 1 General map of the west and south of Peninsular Malaysia, showing sites of Port Dickson coast, Muar river, Johor Bahru coast, Merambong and the Kim Kim river

Activated copper with half spoon had added to each extract to eliminate sulfur linked to the sediments to prevent any influence with the final chromatograms. The extracted sample was decreased by a rotary evaporator, and it was then moved into the top of a chromatography column (5% H_2O-deactivated silica gel) with 60–200 mesh size (Sigma Chemical Company, USA; 9 cm height, 0.9 cm i.d.). First, a pure hexane/DCM combination (3:1, v/v; 20 mL) were used to elute the extracted hydrocarbons, which were then reduced to 1–2 mL. The second stage (4 mL of hexane) was using a column with 0.47 cm i.d., 18 cm height filled with completely activated silica gel to collect the LAB

Table 1 Sampling sites along the west and south of Malaysia

Sample name	Geographical coordination	Location type	Weather condition	Site description
SPD1	N 02° 27′ 882″ E 101° 50.6′ 87″	Beach	Cloudy	Urban and tourism area
SPD2	N 02° 27′ 638″ E 101° 50.7′ 35″	Beach	Cloudy	Urban and tourism area
SPD3	N 02° 28′ 047″ E 101° 50.7′ 96″	Beach	Cloudy	Urban and tourism area
SPD4	N 02° 27′ 943″ E 101° 50.8′ 10″	Beach	Cloudy	Urban and tourism area
SMu1	N 02° 04′ 06.0″ E 102° 33′ 18.1″	River	Cloudy	Urban and industry area
SMu 2	N 02° 03′ 34.9″ E 102° 34′ 27.0″	River	Cloudy	Urban and industry area
SMu 3	N 02° 03′ 18.0″ E 102° 32′ 23.9″	River	Cloudy	Urban and industry area
SJB1	N 01° 27′ 58.2″ E 103° 44′ 30.9″	Coast	Cloudy	Industry and urban area
SJB2	N 01° 27′ 44.1″ E 103° 44′ 84.7″	Coast	Cloudy	Industry and urban area
SJB 3	N 01° 27′ 34.7″ E 103° 45′ 65.9″	Coast	Cloudy	Industry and urban area
SJB4	N 01° 27′ 72.7″ E 103° 46′ 47.2″	Coast	Cloudy	Industry and urban area
SJB5	N 01° 26′ 06.5″ E 103° 55′ 8.40″	Coast	Rainy	Industry and urban area
SMe1	N 01° 22′ 76.0″ E 103° 38′ 08.8″	Island	Sunny	Uninhabited area
SMe2	N 01° 20′ 61.7″ E 103° 36′ 72.0″	Island	Sunny	Uninhabited area
SMe3	N 01° 20′ 40.3″ E 103° 36′ 27.7″	Island	Sunny	Uninhabited area
SKK1	N 01° 27′ 28.2″ E 103° 57′ 76.2″	River	Rainy	Urban and industry area
SKK2	N 01° 26′ 77.6″ E 103° 58′ 04.6″	River	Rainy	Urban and industry area
SKK3	N 01° 25′ 09.6″ E 103° 58′ 3.31″	River	Rainy	Urban and industry area

fraction. The extract was then moved to a 2-mL amber vial and more reduced by a nitrogen gas till near dryness. Prior to gas chromatography-mass spectrometry (GC-MS) measurement, an internal standards with IS = biphenyl-d10 (m/z = 164) had injected into the LAB extract.

A gas chromatograph with a capillary column (DB-5MS) from Agilent Technologies (7890A series) has been used to identify the target chemicals. Briefly, a fused silica capillary column with length of 30 m, 0.25 mm i.d., and a film thickness of 0.25–μm has been employed. Helium, which had a steady pressure (60 kg cm^{-2}) , served as the carrier gas. The mass spectrum data had collected using the selective ion monitoring (SIM) mode. Furthermore, LAB chromatographs had detected at m/z = 91, 92, and 105. The working condition of the GC-MS was set at 70 eV, and the ionization stage had used an external source (200 °C) as well as ~ 1250 eV. The sample injection using splitless mode was then performed with an injection port (300 °C) at a 1-min purge then followed. The temperature of column was maintained at 70 °C for 2 min, 30 °C min^{-1} to 150 °C, and lastly 4 °C min^{-1} to 310 °C for 15 min.

2.3 Method Quality and Assurance

The range of LAB surrogate standards (1-Cn LAB) recovery for all sediment in this study was 87–98% within the allowable percentage (60–120%), showing only a small loss of the targeted chemicals throughout LABs' analytical procedure. A blank sample that contains all the chemicals present in the normal sample had used with every batch of samples (4 samples), to avoid any possible pollution from various ambient sources over the analysis. At fixed volumes, daily freshly manufactured SS, IS, and native standards had spiked into the samples. Additionally, limits of quantitation (LOQ) and detection (LOD) were calculated by determiningthe lowest concentration levels of each calibration curve (Takada and Eganhouse 1998).

2.4 Statistical Analysis

For statistical analyses, IBM®-SPSS 25 software has been employed, and analysis of variance (ANOVA) was performed to demonstrate the significance of the variations in LAB contents and distribution among the sample locations at $p < 0.01$. The Pearson correlation coefficient was employed in the correlation

Table 2 LAB concentration (ng g^{-1} dw) and relative compound ratios in the west and south of Peninsular Malaysia

Compound	Location[a]																	
	Port Dickson				Muar			Johor Bahru coast					Merambong			Kim Kim		
	SPD1	SPD2	SPD3	SPD4	SMu1	SMu2	SMu3	SJB1	SJB2	SJB3	SJB4	SJB5	SMe1	SMe2	SMe3	SKK1	SKK2	SKK3
C_{10}-LABs[b] (ng g^{-1} dw)	17.2	14.9	4.2	6.2	16.4	8.4	8.2	9.9	7.4	15.7	10.3	12.1	7.2	8.8	6.4	9.4	7.7	6.8
C_{11}-LABs (ng g^{-1} dw)	66.7	56.9	19.0	30.7	25.0	14.8	14.2	17.1	12.9	28.4	17.7	19.2	13.1	14.3	11.4	16.3	13.0	12.0
C_{12}-LABs (ng g^{-1} dw)	68.6	61.0	24.0	38.0	41.8	19.7	18.3	27.0	19.1	46.0	27.2	29.5	16.4	19.4	14.1	25.6	20.7	18.6
C_{13}-LABs (ng g^{-1} dw)	86.0	78.4	51.2	63.6	69.4	27.2	25.8	39.1	28.0	69.0	39.6	45.0	23.9	31.9	18.4	41.0	32.6	29.5
C_{14}-LABs (ng g^{-1} dw)	17.4	15.6	13.2	14.8	35.5	21.1	20.9	23.7	20.2	29.5	21.2	25.0	22.2	23.8	17.1	26.8	21.1	21.3
LABs (ng g^{-1} dw)	255.8	226.8	111.6	153.3	188.1	91.2	87.4	116.8	87.6	188.7	115.8	130.7	82.8	98.2	67.4	119.0	95.1	88.2
I/E[c]	4.1	3.7	2.0	2.6	2.2	1.7	1.6	2.0	1.8	2.7	1.9	2.0	1.7	1.7	1.6	1.9	1.8	1.7
L/S[d]	1.6	1.6	1.7	1.6	2.7	2.0	1.9	2.4	2.4	2.7	2.4	2.5	2.00	2.0	2.0	2.4	2.5	2.6
C_{13}/C_{12}[e]	9.7	9.5	13.7	14.3	6.7	4.2	4.2	4.7	4.4	5.7	4.2	6.2	4.9	5.7	4.1	5.1	5.2	5.2
LAB[f] degradation (%)	64	61	40	48	43	33	33	38.	35	49	38	40	34	34	33	38	35	34
TOC[g] (mg g^{-1})	19.7	22.7	14.7	18.9	25.4	22.2	13.8	4.6	19.8	30.9	9.1	3.9	25.4	6.5	35.1	11.5	8.99	81

[a]SPD1, the first letter indicates the station; the second and third letters represent the first letters of location name; the numbers 1, 2, 3, 4 indicate the first, second, third, and fourth station for each location, respectively

[b]C_{10}-LAB, sum of the 26 LAB congeners ranging from 5-C_{10} to 2-C_{10}

[c]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)

[d]L/S, ratio of (5-C_{13}LAB+5-C_{12}LAB) relative to (5-C_{11}LAB+5-C_{10}LAB)

[e]$C_{13}/C_{12,}$ 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 ($r^2 = 0.96$)

[g]TOC (%), total organic carbon

analysis of sampling stations. Regression coefficients efficient were carried out to indicate the potential contribution of multiple sources of LABs.

2.5 TOC Analysis

To find the total organic carbon (TOC) in the sediments, the samples were dried over the night in an oven at 60 °C; then, they were ground using a mortar and pestle. Two milliliters of 1 M HCl was poured to 2 g dried sample until it was completely wet to remove the carbonates. The sediment sample was then dried at 100 °C for 10 h in order to remove the HCl. The TOC % was calculated using an LECO CR-412 Carbon Analyser at 1350 °C and an O_2 boost period of 1 min (Nelson and Sommers 1996). Table 2 shows the calculated TOC percentages.

3 Result and Discussion

3.1 Composition, Distribution, and Concentration LABs

LAB individual congeners are shown in Fig. 2, with form "n-Cm" LAB, where "n" indicates location of phenyl group on the straight alkyl chain and "m" denotes alkyl carbons figure. All of the investigated sediments contained LAB with C_{10}–C_{14}. Table 2 shows LAB concentration ranged from 67.4 and 255.8 ng g^{-1} dw, in samples of the Pulau Merambong as well as Port Dickson coast, respectively.

Figure 3 shows the composition profile of LAB homologs in sampling locations. It observed that the composition of homologs with C_{13} in all study stations has a significant significance for LABs in the examined areas, followed by C_{12} and C_{14}. On the other hand, C_{10} and C_{11} homologs had less level, conforming that those chemicals preferentially degrade through wastewater effluent release. In general, LABs in the rivers and coasts dominated by long-chain LABs (C_{13} and C_{14}), compared to other LAB homologs (C_{10} and C_{11}), suggesting that LABs have long laterally transport in the marine environment. The specific analysis of LAB composition showed that C_{13} homologs were high in the first station of the Port Dickson coast (SPD1), implying that these chemicals pass through anaerobic degradation (Dauner et al., 2015; Alkhadher et al., 2020).

Similarly, C_{10} and C_{11} homologs (short-chain LABs) had low abundance in all sites of this study, suggesting that the most likely explanation in this composition variation of LAB chain lengths is that LABs become more hydrophobic as chain length increases (Sherblom et al., 1992). Additionally, it was observed that C_{10} homolog of this study had less level than those in detergents that had studied by Luo et al. (2008).

The study's findings show that there is a high significant of Pearson connection among the LAB concentrations in the analyzed locations ($p < 0.01$, $r = 0.81$), signifying that LABs could be employed as a tracer of anthropogenic contamination in tropical areas (Table 3). LAB concentrations recorded between 87.4 and 255.8 ng g^{-1} dw, with the first station of the Port Dickson coast (SPD1) recording the highest level and the third station of Pulau Merambong recording the lowest value (SMe3; Fig. 4). It was conforming with a significant difference at $p < 0.01$ of LAB concentration among the study locations (Table 4).

Based on the LAB concentration, the most plausible reason for the spatial dispersion of LABs in the

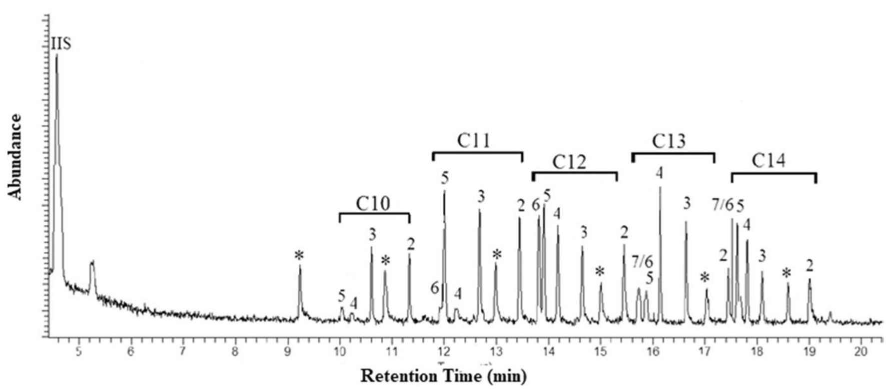
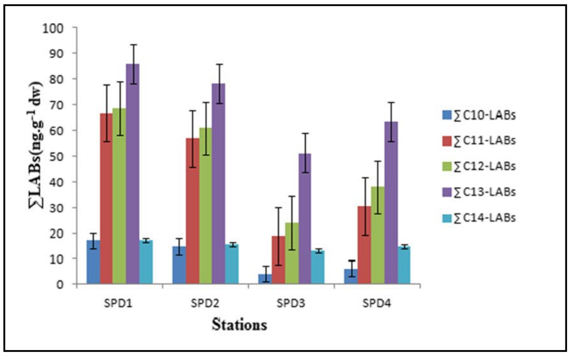
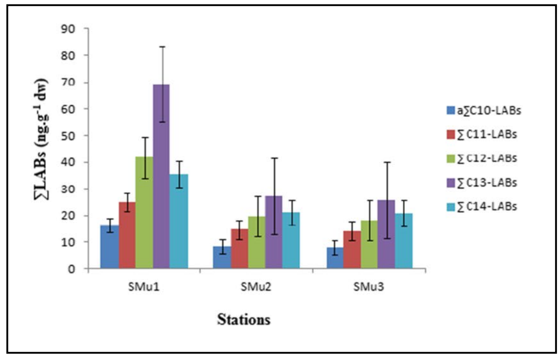


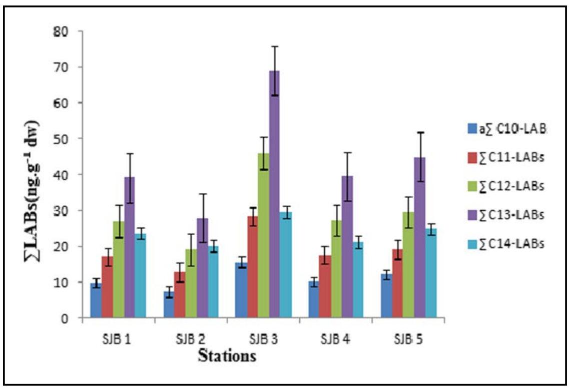
Fig. 2 Gas chromatograms of LABs in surface sediments of the west and south of Peninsular Malaysia. IIS (internal injection standard-biphenyl, d_{10}), surrogates 1-Cn-LABs (n:8-14) from left to right) indicated by asterisks. Subscripts indicate the alkyl chain length. Numbers on the peaks indicate the phenyl substituted position on the alkyl chain



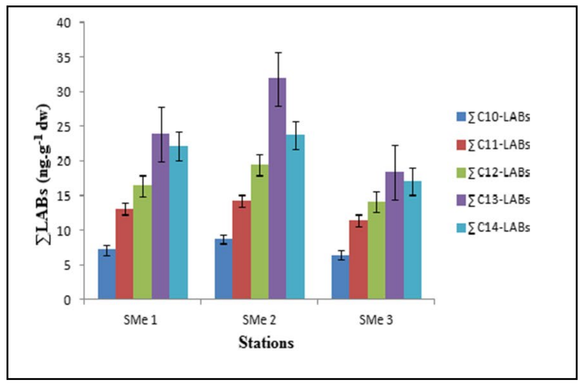
(a) Compositional profiles of LABs in Port Dickson



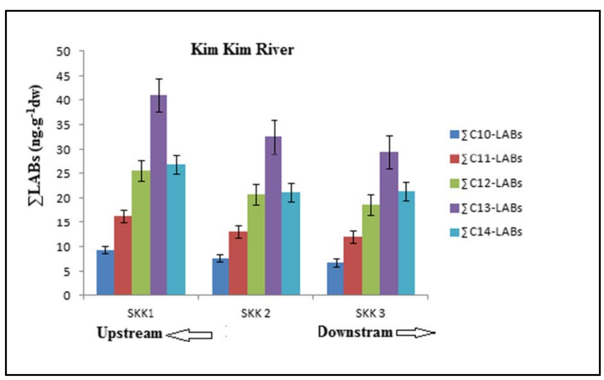
(b) Compositional profiles of LABs in Muar River



(c) Compositional profiles of LABs in Johor Bahru



(d) Compositional profiles of LABs in Merambong



(e) Compositional profiles of LABs in the Kim Kim river

Fig. 3 Compositional profile of LABs in the west and south sediment samples. Standard error bars are shown

investigated areas turned to the discharge of wastewater effluents and their dilution with organic particulate matters that may affect the LABs distribution on the riverine and coastal ecosystems (Zeng et al., 1997; Cabral and Martins, 2018). Instead, a high industrialization, urbanization, and insufficient of sewer system could be behind LABs spatial distribution in those regions (Shahbazi et al., 2010; Sakai et al., 2016).

LAB concentrations in the locations under investigation are significantly less than those in Malacca, Penang estuary, Indonesia, and Anzali wetland (Isobe et al., 2004; Bakhtiari et al., 2018). However,

Table 3 Pearson correlation coefficients between total LAB concentration in the west and south of Peninsular Malaysia. Significant Pearson correlation ($p < 0.01$)

	Location	Total LAB concentration
Pearson correlation	1	− 0.81*
Sig. (2-tailed)		0.01
N	18	18

*Correlation is significant at the 0.01 level (2-tailed

especially in comparison to those observed in the Southern Brazil and Pearl river, LABs were observed with high and comparable levels in this research (Luo et al., 2008; Zacchi et al., 2018).

Obviously, it was found that LAB congeners distributed as such trend: the Port Dickson > Johor Bahru > Muar river > Kim Kim > Pulau Merambong. Thus, it demonstrates that the spatial distribution of LABs in aquatic environments might be affected via the geographical situation of sample stations. Furthermore, it indicated that LABs as an indicator of anthropogenic contamination along the investigated areas relative to those in Malaysia and around the world was low to moderate (Table 5). In addition, it was discovered that the LAB level of this study was high in comparison to those noticed by Isobe et al. (2004).

3.2 TOC Evaluation

Due to of their high hydrophobicity, LABs are more prone to cling to organic matter when they introduce to the aquatic environment. As a result, TOC in the sediments had a relationship with LAB content (Wang et al., 2001).

TOC in the sediment samples was identified, and the relationship between TOC and LABs was assessed by calculating the linear regression and coefficient in this study. TOC recorded from 4.9 in the Johor Bahru (SJB1) with the lowest reading to 81 mg g^{-1} in the Kim Kim (SKK3), with the highest amount of TOC (Table 2). Port Dickson, Muar river, and Merambong island have relationship between LABs and TOC (R^2 = 0.64, 0.54, and − 0.97), respectively (Fig. 5). These

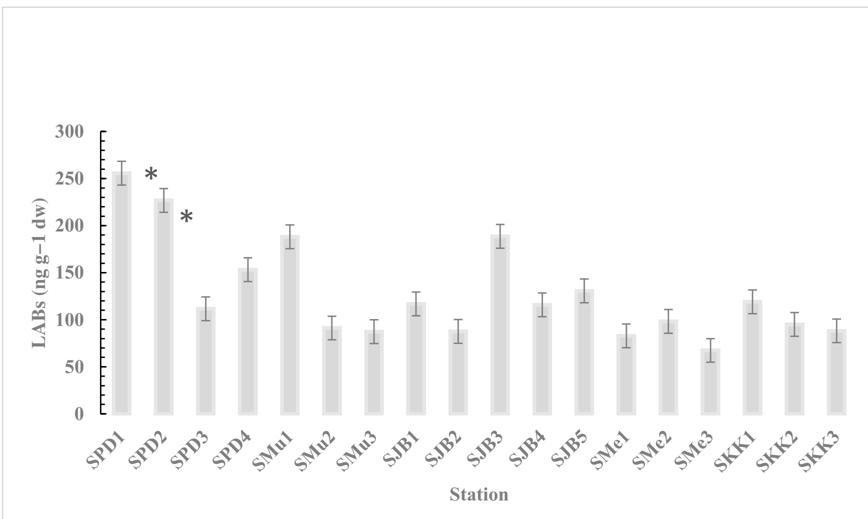


Fig. 4 Concentration of LABs in the west and south sediment samples. Standard error bars are shown

Table 4 Overall analysis of variance for surface sediments of west and south of Malaysia

Source	DF	Sum of square	Mean square	*F* value	Sig.*
Side	1	92,278.39	9227.39	10.11	< .0.01
Locations	5	114,597.32	2291.46	50.20	< .0.01
Error	25	0.00	0.00		
Corrected total	18	143,098.47			

*The mean difference is significant at the 0.01 level results indicate that TOC may be the key factor in the spatial distribution of LAB in those stations. This is in line with the TOC findings from the Dongjiang river, where it was discovered that LAB concentrations were linearly linked with TOC ($R^2 = 0.82$). This result revealed that wastewater discharge was the primary source of organic carbon in the sediments.

However, the Kim Kim and the Johor Bahru shore sediments showed a limited association between LABs and TOC ($R^2 = 0.29$ and -0.42, respectively) (Fig. 5). This shows that TOC was not a decisive factor in LAB distribution and that the main determinant in the redistribution of LABs in those locations was wastewater inputs level from several anthropogenic activities (Fig. 5). Prior researches had noticed that the Selangor and Perak Rivers in Peninsular Malaysia had a weak LABs-TOC relationship with R^2 values of 0.008 and 0.17 (Magam et al., 2015; Masood et al., 2015).

3.3 Evaluation of LAB Degradation and Effluent Treatment's Effectiveness

LAB degradation and sources in aquatic ecosystem surface sediments depends on LAB ratios. In the aquatic environment, I/E ratios are applied to gauge the LABs' degradation and the effectiveness of

Table 5 Total concentrations of ΣLABs from different areas around Malaysia and the world

Location	*N*	Maximum LABs ($ng\ g^{-1}$)[a]	I/E ratio[b]	Degradation[c] (%)	Reference
South Atlantic Estuary	15	210	2.5	47	Cabral and Martins (2018)
Southern Brazil	3	15.3	1.4	27	Zacchi et al. (2018)
Humber Estuary and Wash, UK	18	84.8	2.1	41	Raymundo and Preston (1992)
Anzali Wetland, Iran	167	109,000	1.3	24	Bakhtiari et al. (2018)
Malacca, Malaysia	1	1080	2.0	39	Isobe et al. (2004)
Muar River, Malaysia	1	32	2.8	51	Isobe et al. (2004)
Penang Estuary, Malaysia	1	3000	1.5	29	Isobe et al. (2004)
Prai River, Malaysia	1	25	3.4	58	Isobe et al. (2004)
Kim Kim River, Malaysia	1	122	1.8	36	Isobe et al. (2004)
Kim Kim Estuary, Malaysia	1	6	1.2	21	Isobe et al. (2004)
Nibong Tebal, Malaysia	1	168	2.1	41	Isobe et al. (2004)
Indonesia	20	42,600	2.1	41	Isobe et al. (2004)
Sarawak River, Malaysia	9	7390	1.0	15	Magam et al. (2012)
Sembulan River, Malaysia	6	5570	1.8	36	Magam et al. (2012)
Zhujiang River	11	2330	1.5	29	Luo et al. (2008)
Dongjiang River	10	566	1.9	38	Luo et al. (2008)
Xijiang River	8	69.4	1	15	Luo et al. (2008)
Pearl River Estuary	8	26	1.5	29	Luo et al. (2008)
South China Sea	28	23	0.9	11	Luo et al. (2008)
The Pearl River Delta	96	11,200	1.7	34	Ni et al. (2008)
Santos Bay, Brazil	14	117	2.9	55	Martins et al. (2008)
Dongjiang River	45	410	1.4	27	Zhang et al. (2012)
Outfalls of paper mills	3	3270	1.3	24	Zhang et al. (2012)
Jakarta Bay	7	86,800	0.9	12	Rinawati et al. (2012)
Tokyo Bay	2	1110	2.8	51	Rinawati et al. (2012)
Detergents	10	5,300,000	1.7	34	Raymundo and Preston (1992)

N the number of samples

[a]LAB = sum of concentrations of all secondary LAB congeners having C10–C14 alkyl chain

[b]I/E = (6_C12+5_C12)/4_C12+3_C12 + 2_C12)

[c]LAB deg =81*log (I/E ratio) +15 ($r^2 = 0.96$)

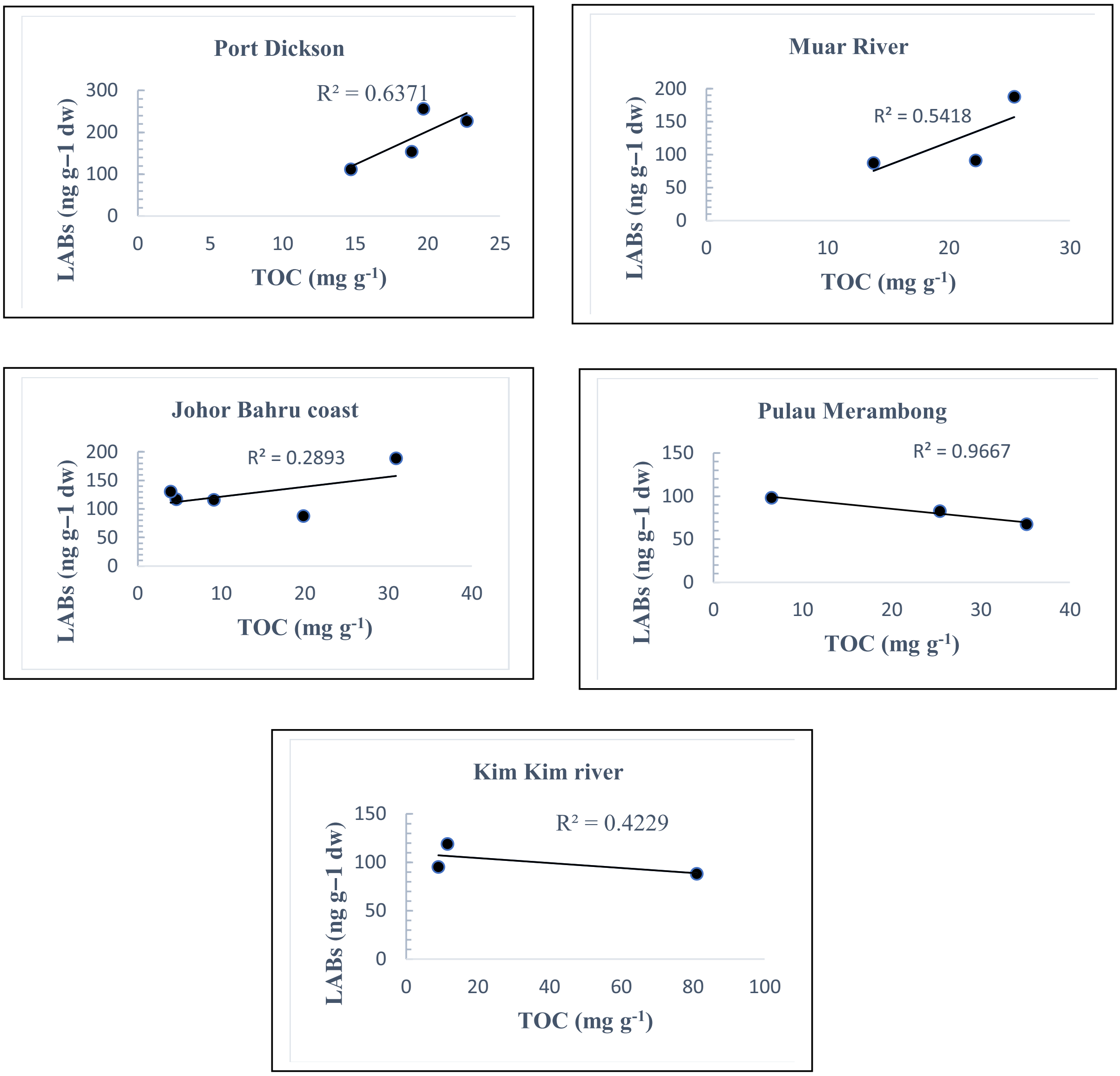


Fig. 5 Scatter plots of LABs and TOC in sediment samples of the west and south sediment samples

wastewater treatments (Takada and Ishiwatari, 1990, Alkhadher et al., 2021).

LAB degradation in the surface sediments of studied areas varied in the range from low (33%) at SMu2, SMu3, and SMe3 of Muar river and Pulau Merambong to high (64, 61%) at SPD1 and SPD2 of Port Dickson coast, showing that Port Dickson's coast has the greatest rate of LAB biodegradation compared to other sampling sites in Peninsular Malaysia (see Table 2).

Figure 6 shows that the first station of Port Dickson (SPD1) has the highest I/E ratio with 4.1 and the stations SMu2, SMu3, SMe1, SMe2, SMe3, and SKK3 of the Muar, Pulau Merambong, and Kim Kim have the lowest I/E ratios, indicating that most of LABs in studied areas came from secondary and primary treatment sources. This explained the presence of many STPs in Peninsular Malaysia.

The results showed that the Kim Kim, Pulau Merambong, and most stations of the Muar have higher

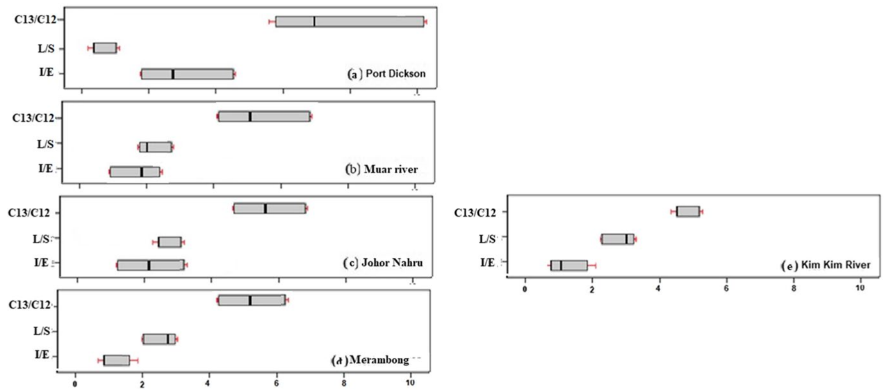


Fig. 6 Values of the C_{13}/C_{12}, L/S, and I/E ratios in the samples of the west and south sediment samples

concentrations of primary wastewater effluents (SMu2 and Smu3) and the Johor Bahru (SJB2 and SJB4), while the secondary wastewater effluents discharged into the Port Dickson coast, the Muar station (SMu1), and Johor Bahru stations (SJB1, SJB3, and SJB5). Multiple sources emit LABs into the environment, for example, wastewater from cities or industries. Wastewater particles have a strong attachment to organic matters and sewage particles and may therefore improve the transfer of LASs between their originating sources and eventual transportation into the oceans (Zhang et al., 2012; Harwood, 2014; Cabral et al., 2018).

Furthermore, ratios of L/S and C_{13}/C_{12}, determined to more assessment of LAB degradation in studied areas. L/S ratios in this study ranged from 1.6 at SPD1, SPD2, and SPD4 of Port Dickson coast to 2.6 at SKK3 of the Kim Kim river, and they had more than 1.8 that was detected in detergents (Ni et al., 2008), representing improvement of LAB biodegradation. Furthermore, C_{13}/C_{12} ratio ranged from 4.1 at the third station of Pulau Merambong (SMe3) to 14.3 at the fourth station of the Port Dickson station (SPD4), and this ratio was more than 1.7 found in the sediments of coastal areas (Liu et al., 2013).

In recent years, there has been a significant direct discharge from ferries and boats due to the fact that locals use the river for recreational and fishing purposes (Abu Samah et al., 2011). As a result, detergent residues and boat wash cause an increase in LABs in the sediments, which drastically affects molecular indices. In developing countries like Malaysia, it is evident that raw, primary, and additional secondary processed effluents discharged into the aquatic environment comprise LAB inputs. However, proximity to places with high population densities, main industrial and tourism-related activities can result in a rise in LAB concentration and a decrease in I/E ratios in certain locales. Instead, the locations with low urbanized, industrialized, and population close to the river causing in low concentration in the Pulau Merambong and the Kim Kim compared to the other sampling locations.

4 Conclusion

LAB concentrations with significant differences in the sediments of the analyzed places of the west and south of Peninsular Malaysia were 67.4–255.8 ng g^{-1} dw. The Port Dickson coast has the highest LABs than other sampling locations. Compared to LC- ones, SC-homologs are less prevalent, with the lowest levels in the Pulau Merambong. The results of I/E ratios demonstrated that the majority of LABs are from the primary treatment effluents, whereas secondary treatment was the predominant source along the Port Dickson coast. A large LAB content could be caused by insufficient STPs, which might not be able to serve a high population in the studied. As a result, the study's results revealed that as the population increase, untreated industrial and residential wastewater concerns could remain for a very long time. It is expected that the level of industrial waste and municipal wastewater flow into Malaysia's coasts and rivers would rise in the near future. As a result, it is crucial to take the appropriate actions to upgrade the wastewater treatment system. In light of this, routine wastewater pollution assessments should be performed in addition to sewage system improvements to lessen environmental concerns and promote public health in riverine and coastal areas.

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Data Availability The metadata used to support the findings of this study have been deposited in the University 301 Putra Malaysia repository at http://upm.edu.my/.

Declarations

Conflict of Interest The authors declare that they have no conflict of interest

References

Abu Samah, B., Yassin, S. M., Shaffril, H. A. M., Hassan, M. S., Othman, M. S., Abu Samah, A., & Ramli, S. A. (2011). Relationship to the river: The case of the Muar River community. *American Journal of Environmental Sciences, 7*(4), 362–369.

Alkhadher, S. A. A., Zakaria, M. P., Yusoff, F. M., Kannan, N., Suratman, S., Keshavarzifard, M., Magam, S. M., Masood, N., Vaezzadeh, V., & Sani, M. S. A. (2015). Baseline distribution and sources of linear alkylbenzenes (LABs) in surface sediments from Brunei Bay, Brunei. *Marine Pollution Bulletin, 101*(1), 397–403.

Alkhadher, S. A. A., Zakaria, M. P., Yusoff, F. M., Kannan, N., Suratman, S., Magam, S. M., Masood, N., Keshavarzifard, M., Vaezzadeh, V., & Sani, M. S. A. (2016). Distribution and sources of linear alkylbenzenes (LABs) in surface sediments from Johor Bahru Coast and the Kim Kim River, Malaysia. *Environmental Forensics, 17*(1), 36–47.

Alkhadher, S. A. A., Kadir, A. A., Zakaria, M. P., Al-Gheethi, A., Magam, S. M., & Masood, N. (2020). Monitoring of sewage pollution in the surface sediments of the coastal ecosystems using linear alkylbenzenes (LABs) as molecular markers. *Journal of Soils and Sediments, 20*, 3230–3242.

Alkhadher, S. A. A., Kadir, A. A., Zakaria, M. P., Al-Gheethi, A., Keshavarzifard, M., Masood, N., Alenezi, K. M., & Magam, S. M. (2020). Linear alkylbenzenes in surface sediments of an estuarine and marine environment in Peninsular Malaysia. *Marine Pollution Bulletin, 153*, 111013.

Alkhadher, S. A. A., Kadir, A. A., Zakaria, M. P., Al-Gheethi, A., & Asghar, B. H. M. (2020). Determination of linear alkylbenzenes (LABs) in mangrove ecosystems using the oyster Crassostrea belcheri as a biosensor. *Marine Pollution Bulletin, 154*, 111115.

Alkhadher, S. A. A., Zakariab, M., Suratman, S., Alanazi, T. Y. A., Al-Bagawi, A. H., Magam, S. M., Masood, N., Kadir, A. A., & Al-Gheethi, A. (2021). Assessment of Sewage Molecular Markers in Port Dickson Coast and Kim Kim River with Sediment Linear Alkylbenzenes. *Polycyclic Aromatic Compounds*. https://doi.org/10.1080/10406638.2022.2039234

Bakhtiari, A. R., Javedankherad, I., Mohammadi, J., & Taghizadeh, R. (2018). Distribution of linear alkylbenzenes as a domestic sewage molecular marker in surface sediments of International Anzali Wetland in the southwest of the Caspian Sea, Iran. *Environmental Science and Pollution Research, 25*(12). https://doi.org/10.1007/s11356-018-1942-1

Cabral, A. C., & Martins, C. C. (2018). Insights about sources, distribution, and degradation of sewage and biogenic molecular markers in surficial sediments and suspended particulate matter from a human-impacted subtropical estuary. *Environmental Pollution, 241*, 1071–1081.

Cabral, A. C., Stark, J. S., Kolm, H. E., & Martins, C. C. (2018). An integrated evaluation of some faecal indicator bacteria (FIB) and chemical markers as potential tools for monitoring sewage contamination in subtropical estuaries. *Environmental Pollution, 241*, 739–749.

Cabral, A. C., Dauner, A. L. L., Xavier, F. C. B., Garcia, M. R. D., Wilhelm, M. M., Santos, V. C. G., Netto, S. A., & Martins, C. C. (2020). Tracking the sources of allochthonous organic matter along a subtropical fluvial-estuarine gradient using molecular proxies in view of land uses. *Chemosphere, 251*(2020), 126435.

Chaler, R., Cantón, L., Vaquero, M., & Grimalt, J. O. (2004). Identification and quantification of n-octyl esters of alkanoic and hexanedioic acids and phthalates as urban wastewater markers in biota and sediments from estuarine areas. *Journal of Chromatography A, 1046*(1-2), 203–210.

Dauner, A. L. I., Hernández, E. A., MacCormack, W. P., & Martins, C. C. (2015). Molecular characterisation of anthropogenic sources of sedimentary organic matter from Potter Cove, King George Island, Antarctica. *Science of the Total Environment, 502*, 408–416.

Hagemann, L., Buchty-Lemke, M., Lehmkuhl, F., Alzer, J., Kümmerle, E. A., & Schwarzbauer, J. (2018). Exhaustive screening of long-term pollutants in riverbank sediments of the Wurm River, Germany. *Water, Air, & Soil Pollution, 229*, 197.

Halpern, B. S., et al. (2008). A global map of human impact on marine ecosystems. *Science, 319*, 948–952.

Huang, Y., Liu, L., Liu, G., & Gong, Y. (2021). Occurrence and fate of linear alkylbenzenes and their potential as environmental molecular markers in highly urbanized river systems. *Science of the Total Environment, 760*(2021), 14394.

Harwood, J. J. (2014). Molecular markers for identifying municipal, domestic and agricultural sources of organic matter in natural waters. *Chemosphere, 95*(2014), 3–8.

Ibrahim, Z. Z. (1988). *An integrated approach to the investigation of an equatorial estuary: the Klang River Estuary*. University of Southampton.

Isobe, K. O., Tarao, M., Zakaria, M. P., Chiem, N. H., Minh, L. Y., & Takada, H. (2002). Quantitative application of fecal sterols using gas chromatography-mass spectrometry to investigate fecal pollution in tropical waters: Western Malaysia and Mekong Delta, Vietnam. *Environmental Science & Technology, 36*(21), 4497–4507.

Isobe, K. O., Zakaria, M. P., Chiem, N. H., Minh, L. Y., Prudente, M., Boonyatumanond, R., Saha, M., Sarkar, M., &

Takada, H. (2004). Distribution of linear alkylbenzenes (LABs) in riverine and coastal environments in South and Southeast Asia. *Water Research, 38*(9), 2449–2459.

Liu, L. Y., Wang, J. Z., Wong, C. S., Qiu, J. W., & Zeng, E. Y. (2013). Application of multiplegeochemical markers to investigate organic pollution in a dynamic coastal zone. *Environmental Toxicology and Chemistry, 32*, 312–319.

Luo, X. J., Chen, S. J., Ni, H. G., Yu, M., & Mai, B. X. (2008). tracing sewage pollution in the Pearl River Delta and its adjacent coastal area of South China Sea using linear alkylbenzenes (LABs). *Marine Pollution Bulletin, 56*(1), 158–162.

Magam, S. M., Zakaria, M. P., Halimoon, N., Masood, N., & Alsalahi, M. A. (2012). Aliphatic distribution of linear alkylbenzenes (LABs) in sediments of Sarawak and Sembulan rivers, Malaysia. *EnvironmentAsia, 5*(1), 48–55.

Magam, S. M., Halimoon, N., Zakaria, M. P., Aris, A. Z., Kannan, N., & Masood, N. (2015). Evaluation of distribution and sources of sewage molecular marker (LABs) in selected rivers and estuaries of Peninsular Malaysia. *Environmental Science and Pollution Research, 23*(6), 5693–5704.

Martins, C. C., Ferreira, J. A., Taniguchi, S., Mahiques, M. M., Bícego, M. C., & Montone, R. C. (2008). Spatial distribution of sedimentary linear alkylbenzenes and faecal steroids of Santos Bay and adjoining continental shelf, SW Atlantic, Brazil: origin and fate of sewage contamination in the shallow coastal environment. *Marine Pollution Bulletin, 56*(7), 1359–1363.

Masood, N., Zakaria, M. P., Halimoon, N., Aris, A. Z., Magam, S. M., Kannan, N., Mustafa, S., Ali, M. M., Keshavarzifard, M., Vaezzadeh, V., Alkhadher, S. A. A., & Al-Odaini, N. A. (2015). Anthropogenic waste indicators (AWI) particularly PAHs and LABs in Malaysian sediments: application of aquatic environment for identifying anthropogenic pollution. *Marine Pollution Bulletin, 102*(1), 160–175.

Masood, N., Alkhadher, S. A. A., Magam, S. M., Halimoon, N., Alsukaibi, A., Zakaria, M. P., Vaezzadeh, V., Keshavarzifard, M., Maisara, S., & Break, M. K. (2021). Monitoring of linear alkyl benzenes (LABs) in riverine and estuarine sediments in Malaysia. *Environmental Geochemistry and Health volume, 44*, 3687–3702.

Melo, M. G., Silva, B. A., Costa, G. S., Neto, J. C. A. S., Soares, P. K., Val, A. L., Chaar, J. S., Koolen, H. H. F., & Bataglion, G. A. (2019). Sewage contamination of Amazon streams crossing Manaus (Brazil) by sterol biomarkers. *Environmental Pollution, 244*, 818–826.

Nelson, D., & Sommers, L. (1996). Total carbon, organic carbon and organic matter. In D. L. Sparks (Ed.), *In 'Methods of soil analysis. Part 3: Chemical methods'* (pp. 961–1010). Soil Science Society of America.

Ni, H. G., Lu, F. H., Wang, J. Z., Guan, Y. F., Luo, X. L., & Zeng, E. Y. (2008). Linear alkylbenzenes in riverine runoff of the Pearl River Delta (China) and their application as anthropogenic molecular markers in coastal environments. *Environmental Pollution, 154*(2), 348–355.

Oller, I., Malato, S., & Sánchez-Pérez, J. A. (2011). Combination of advanced oxidation processes and biological treatments for wastewater decontamination, A review. *Science of the Total Environment, 409*, 4141–4166.

Raymundo, C., & Preston, M. (1992). The distribution of linear alkylbenzenes in coastal and estuarine sediments of the Western North Sea. *Marine Pollution Bulletin, 24*(3), 138–146.

Ricking, M., Schwarzbauer, J., & Franke, S. (2003). Molecular markers of anthropogenic activity in sediments of the Havel and Spree Rivers (Germany). *Water Research, 37*(2003), 2607.

Rinawati Koike, T., Koike, H., Kurumisawa, R., Ito, M., Sakurai, S., Togo, A., Saha, M., Arifinc, Z., & Takada, H. (2012). Distribution, source identification, and historical trends of organic micro pollutants in coastal sediment in Jakarta Bay, Indonesia. *Journal of Hazardous Materials, 217*, 208–216.

Sakai, N., Yusof, R., Sapar, M., Yoneda, M., & Ali, M. M. (2016). Spatial analysis and source profiling of beta-agonists and sulfonamides in Langat River basin, Malaysia. *Science of the Total Environment, 548-549*, 43–50.

Sakai, N., Shirasaka, J., Matsui, Y., Ramli, M. R., Yoshida, K., Mohd, M. A., & Yoneda, M. (2017). Occurrence, fate and environmental risk of linear alkylbenzene sulfonate in the Langat and Selangor River basins, Malaysia. *Chemosphere, 172*(2017), 234–241.

Shahbazi, A., Zakaria, M. P., Yap, C. K., Tan, S. G., & Surif, S. (2010). Use of different tissues of Perna viridis as biomonitors of polycyclic aromatic hydrocarbons (PAHs) in the coastal waters of Peninsular Malaysia. *Environmental Forensics, 11*(3), 248–263.

Sherblom, P. M., Gschwend, P. M., & Eganhouse, R. P. (1992). Aqueous solubilities, vapor pressures, and 1-octanol-water partition coefficients for C9-C14 linear alkylbenzenes. *Journal of Chemical & Engineering Data, 37*(4), 394–399.

Takada, H., & Ishiwatari, R. (1990). Biodegradation experiments of linear alkylbenzenes (LABs): isomeric composition of C12 LABs as an indicator of the degree of LAB degradation in the aquatic environment. *Environmental Science & Technology, 24*(1), 86–91.

Takada, H., & Eganhouse, R. P. (1998). Molecular markers of anthropogenic waste: Their use in determining sources, transport pathways and fate of wastes in the environment, In. *Encyclopedia of Environmental Analysis and Remediation, 5*, 2883–2940.

Thomes, M. T., Vaezzadeh, V., Zakaria, M. P., & Bong, C. W. (2019). Use of sterols and linear alkylbenzenes as molecular markers of sewage pollution in Southeast Asia. *Environmental Science and Pollution Research, 26*(2019), 31555–31580.

Tsutsumi, S., Yamaguchi, Y., Nishida, I., Akiyama, K., Zakaria, M. P., & Takada, H. (2002). Alkylbenzenes in mussels from South and South East Asian coasts as a molecular tool to assess sewage impact. *Marine Pollution Bulletin, 45*(1), 325–331.

Wang, X. C., Zhang, Y. X., & Chen, R. F. (2001). Distribution and partitioning of polycyclic aromatic hydrocarbons (PAHs) in different size fractions in sediments from Boston Harbor, United States. *Marine Pollution Bulletin, 42*(11), 1139–1149.

Zacchi, F. L., Flores-Nunes, F., Mattos, J. J., Lima, D., Lüchmann, K. H., Sasaki, S. T., Bícego, M. C., Satie Taniguchi, S., Montone, R. C., Almeida, E. A., & Bainy, A. C. D. (2018). Biochemical and molecular responses in oysters Crassostrea brasiliana collected from estuarine aquaculture areas in Southern Brazil. *Marine Pollution Bulletin, 135*, 110–118.

Zakaria, M. P., Takada, H., Tsutsumi, S., Ohno, K., Yamada, J., Kouno, E., & Kumata, H. (2002). Distribution of polycyclic aromatic hydrocarbons (PAHs) in rivers and estuaries in Malaysia: a widespread input of petrogenic PAHs. *Environmental Science & Technology, 36*(9), 1907–1918.

Zeng, E. Y., Khan, A. R., & Tran, K. (1997). Organic pollutants in the coastal environment Off San Diego, California. 3. Using linear alkylbenzenes to trace sewage-derived organic materials. *Environmental Toxicology and Chemistry, 16*(2), 196–201.

Zhang, K., Wang, J. Z., Liang, B., Shen, R. L., & Zeng, E. Y. (2012). Assessment of aquatic wastewater pollution in a highly industrialized zone with sediment linear alkylbenzenes. *Environmental Toxicology and Chemistry, 31*(4), 724–730.

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