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Baseline distribution and sources of linear alkyl benzenes (LABs) in surface sediments from Brunei Bay, Brunei

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ABSTRACT

Sewage pollution is one of major concerns of coastal and shoreline settlements in Southeast Asia, especially Brunei. The distribution and sources of LABs as sewage molecular markers were evaluated in surface sediments collected from Brunei Bay. The samples were extracted, fractionated and analyzed using gas chromatography-mass spectrometry (GC-MS). LABs concentrations ranged from 7.1 to 41.3 ng g^{-1} dry weight (dw) in surficial sediments from Brunei Bay. The study results showed LABs concentrations variably due to the LABs intensity and anthropogenic influence along Brunei Bay in recent years. The ratio of Internal to External isomers (I/E ratio) of LABs in sediment samples from Brunei Bay ranged from 0.56 to 2.17 along Brunei Bay stations, indicating that the study areas were receiving primary and secondary effluents. This is the first study carried out to assess the distribution and sources of LABs in surface sediments from Brunei Bay, Brunei.

Discharge and emission of sewage contamination are derived from human activities such as industrial development, urbanization, tourism, etc. The control of aquatic pollution has been identified as an immediate need for sustained management and conservation of the existing fisheries and aquatic resources (Islam and Tanaka, 2004). By far, sewage is the greatest volume of waste discharged to the marine environment. Highly populated cities generate huge loads of such wastes daily which are finally washed out by the drainage systems that generally release into nearby rivers or aquatic systems. Sewage contamination can be assessed by microbiological and chemical markers (Vivian, 1986; Takada and Eganhouse, 1998).

Linear alkylbenzenes (LABs) are one group of the chemical markers which have been successfully utilized as organic molecular markers for evaluating the source of sewage pollution (Eganhouse, 1997). Owing to their source specificity, resistance to degradation and persistence in marine sediments for a long time, molecular markers such as LABs have been important for studying anthropogenically derived organic matter input and its impact on aquatic environments (Takada and Eganhouse, 1998). Due to its improved biodegradability and cost-effectiveness, LABs have completely replaced the older branched alkylbenzene in the production of surfactants that have been used in household laundry detergents and dishwashing applications since the 1960s. LABs have isomers with different phenyl-substitution positions on the alkyl chains. It is easier to biodegrade external isomers (isomers whose phenyl substitution positions are close to the terminal end of the alkyl chain) than internal isomers (isomers whose substitution positions are close to the center of the alkyl chain). Thus, the distribution of LAB isomers indicates the level of LAB biodegradation (Takada and Ishiwatari, 1990). Furthermore, the isomeric structure and concentration of LABs reflect the magnitude and types of sewage discharged into the aquatic environment, such as raw sewage versus secondary effluents (Tsutsumi et al., 2002).

The I/E ratio (a ratio of the total of Internal to External isomers) has been proposed as an indicator of LAB degradation level in an aquatic environment (Takada and Ishiwatari, 1990). Because of these attributes, LABs are good indicators of human activities associated with sewage contamination in different regions around the world (Eganhouse et al., 1983; Takada et al., 1992; Isobe et al., 2004; Medeiros and Bícego, 2004; Luo et al., 2008; Martins et al., 2008; Ni et al., 2009; Venkatesan et al., 2010; Martins et al., 2012; Rinawati et al., 2012; Alkhadher et al., in press). All kinds of pollution stemming from human activities will ultimately settle down in surface sediments (Abdullah et al., 1999).

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Most of the contaminants leave their fingerprints in sediments, thus sediment analysis offers certain advantages compared to water analysis.

Brunei is a Southeast Asian country consisting of two unconnected parts with a total area of 5.8 km2 on the island of Borneo and the population of Brunei is 415,000 people of which 76% live in urban areas. The rate of urbanization is estimated at 2.13% per year from 2010 to 2015 (Oxford Business Group, 2013).

The purpose of this study is to evaluate source of sewage pollution by applying LABs and I/E ratios in the surface sediments from aquatic environment of the Brunei Bay. This study provides valuable insights into the degradation of LABs in the marine environment as well as act as a gauge for the efficiency of sewage treatment plants (STPs) in this area.

The present study was conducted in the Brunei Bay which is located on the northwestern coast of Borneo Island. Surface sediment samples were collected from fourteen locations at Brunei Bay during December 2013 (Fig. 1, Table 1). The sediments were sampled with a stainless steel Ekman dredge sampler. The top 4 cm of surface sediments was placed into a pre-cleaned plastic ziplock bag and stored at −20 °C. The sediments were then homogenized and freeze-dried.

Purification and fractionation of the sediment samples was performed using a method presented by Zakaria et al. (2002). Briefly, the previously homogenized, freeze-dried and precisely weighed samples were soxhlet-extracted using 250 mL of high purity dichloromethane (DCM) for 10 h. Prior to each sample extraction, the 1-Cn LAB (50 μL) were added as surrogate standards where 1- refers to the first isomer of each LAB homolog, n refers to carbon number within the range of 8 to 14. The extraction of the samples was followed by activated copper treatment to remove elemental sulfur. The extracts were rotary-evaporated to near dryness and transferred onto the top of 5% H_2O deactivated silica gel (60–200 mesh size, Sigma Chemical Company, USA) in a glass chromatographic column (0.9 cm i.d. and 9 cm height). Exactly 20 mL of high purity Hexane/DCM (3:1, v/v) was used as an elution solvent for hydrocarbon fraction. The extracts were rotary evaporated and reduced to 1–2 mL and sequentially fractionated with a fully activated silica gel column (0.47 cm i.d. and 18 cm height) to get LAB fractions using 4 mL of high purity hexane. The LAB fractions were then transferred to a 2 mL amber vial and evaporated to near dryness using a gentle stream of nitrogen. The internal standards (biphenyl-d10, m/z = 164) contained in 10 ppm internal injection was added to each blank and sample extract before instrumental analyses. The instrumental procedure involved the use of a GC–MS Agilent model with a 5MS fused-silica capillary column (30 m by 0.25 mm i.d. and 0.25-μm film thickness) to analyze the LABs.

The 26 congeners of LABs were analyzed. A 1-μL aliquot of purified samples was introduced into the GC–MS injector. The GC–MS temperature program started at 70 °C for 2 min, with a ramp of 30 °C per minute until 150 °C. The temperature was further increased to 310 °C with an increasing rate of 4 °C/min for 15 min. The analysis was done using selected ion monitoring (SIM) mode with splitless injection. Individual LABs were monitored in SIM mode at (mass/charge ratio) m/z = 91, 92 and 105. The oven temperature was held at 70 °C for 2 min, then continued at 30 °C/min to 150 °C, 5 °C/min to 310 °C and held for 6 min.

The mass spectrometer was scanned repeatedly at an ionization potential of 70 eV with the source at 200 °C and electron multiplier voltage at ~2000 eV.

Quantification of target compounds of LABs was carried out based on external calibration curves using a series of LAB standard mixture solutions for the 26 target compounds with different concentrations (0.25, 0.5, 1, 2.5 and 5 mg/L) were prepared. Determination of target compounds was achieved based on matching their compound ionization and retention times with the standard mixture of LABs. All of the LABs in sediment samples were calculated based on dry weight (dw).

Quality assurance and quality control were considered while conducting the analytical processes. The reasonable efficiency of surrogate recovery (1-Cn LAB) for LABs indicates that there is a minimal possibility of loss of target compounds during analysis due to their non-volatile, non-polar nature. Ranges of recoveries of the LABs surrogates

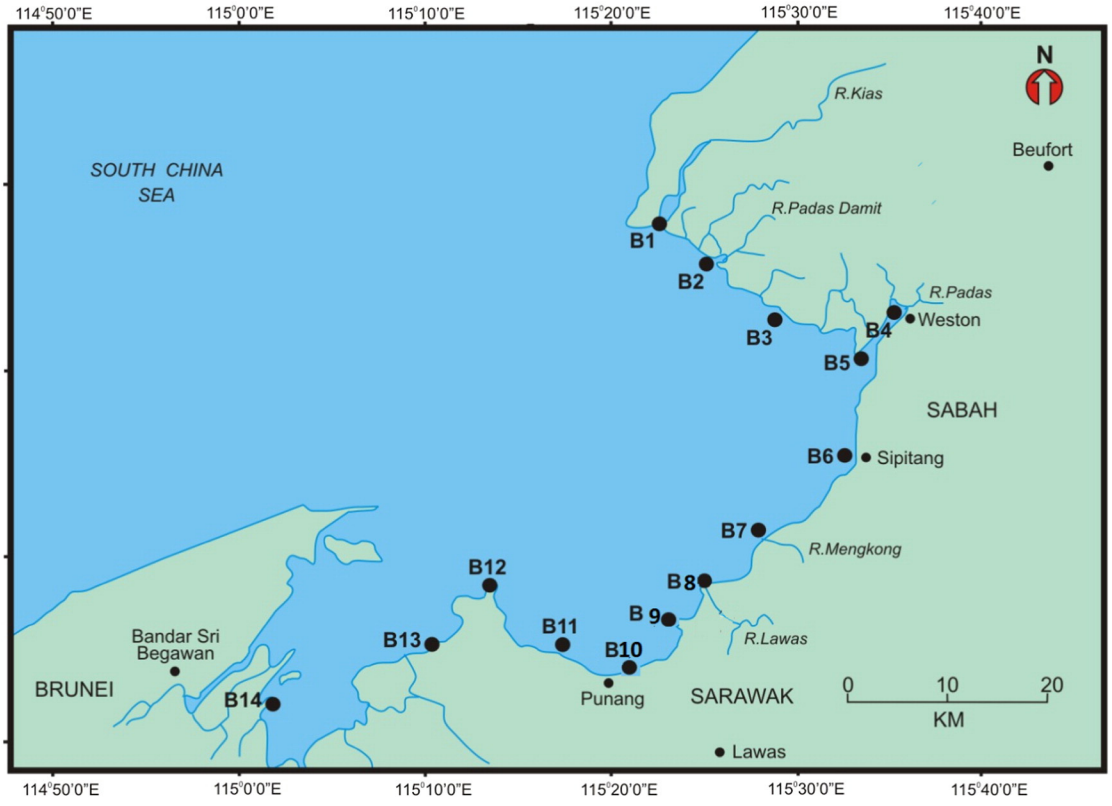

Fig. 1. Sampling stations along Brunei Bay (B1–B14).

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Table 1
Description of sampling site along the selected areas of Brunei Bay.

No.	Station	Geographical coordination	Sediment sample	Average depth	Sampling date
1	B 1	N 5°17′39.12″ E 115°22′38.7588″	Bay	1–2 m	12/12/2013
2	B 2	N 5°15′30.8988″ E 115°24′58.7016″	Bay	1–2 m	12/12/2013
3	B 3	N 5°12′36.3996″ E 115°28′48.3996″	Bay	1–2 m	12/12/2013
4	B 4	N 5°12′54.1008″ E 115°35′15.6012″	Bay	1–2 m	12/12/2013
5	B 5	N 5°10′26.04″ E 115°33′32.4″	Bay	1–2 m	12/12/2013
6	B 6	N 5°5′11.7996″ E 115°32′43.4004″	Bay	1–2 m	12/12/2013
7	B 7	N5°1′53.0004″ E115°29′ 44.9988″	Bay	1–2 m	12/12/2013
8	B 8	N 4°56′40.2″ E 115°26′7.1016″	Bay	1–2 m	12/12/2013
9	B 9	N 4°58′40.1016″ E 115°24′55.4004″	Bay	1–2 m	12/12/2013
10	B 10	N 4°54′43.8984″ E 115°21′50.4″	Bay	1–2 m	12/12/2013
11	B 11	N 4°55′11.3016″ E 115°17′29.7996″	Bay	1–2 m	12/12/2013
12	B 12	N 4°58′16.2984″E 115°13′40.1016″	Bay	1–2 m	12/12/2013
13	B 13	N 4°55′12.36″ E 115°10′24.888″	Bay	1–2 m	12/12/2013
14	B 14	N 4°5′54″ E 115°1′55.2″	Bay	1–2 m	12/12/2013

were 82–104%, throughout the sample analyses. Each sample of LAB concentration obtained was recovery-corrected against the surrogate internal standards (1-Cn LABs) spiked. Procedural blanks for LABs were conducted with each batch of samples together with the samples from extraction up to instrumental analysis, to determine any cross-contamination during the analytical procedures and from glassware. All standards were purchased from Sigma Chemical Company, USA and freshly prepared prior to the actual analysis. In order to test the GC–MS condition, auto tuning was performed on a daily basis.

Total organic carbon (TOC; Table 2) measurements for the sediment samples were conducted with LECO CR-412 analyzer. The TOC analytical procedures were similar to those described by Bakhtiari et al. (2010) and Raza et al. (2011). The sediment samples were dried and then ground and homogenized using mortar and pestle. About 1–2 g of each sediment sample was weighed and 1–2 mL of 1 M hydrochloric acid (HCl) was added to remove inorganic carbon (carbonates) until the sample was totally moist with HCl. Then the samples were dried at 100 °C for 10 h to remove HCL. An aliquot of each sample was reweighed and analyzed using LECO CR-412 analyzer at 135 °C for 60 s to measure the TOC percentage. The TOC is presented in mg/g unit.

The 26 congeners of LABs were detected in all samples of Brunei Bay. There has been no similar large-scale investigation in Brunei Bay and this study is considered the first study carried out on the characterization and distribution of LABs. The total LAB concentrations in surface sediments from Brunei Bay ranged from 7.1 to 41.3 ng g^{-1} dw with arithmetic mean of 19.8 ng g^{-1} dw (Table 2). However, the total concentrations of LABs in the Brunei Bay were much lower than those in the Northern Tokyo Bay, India, Indonesia and Chaohu Lake (Takada et al., 1992; Isobe et al., 2004; Wang et al., 2012). On the other hand, LAB concentrations were also comparable or greater than those in the Xijiang River, Pearl River Estuary, the Admiralty Bay, Brazil, continental shelf of China and Potter Cove and vicinity, Antarctica (Luo et al., 2008; Martins et al., 2012; Wei et al., 2014; Dauner et al., 2015). The comparison of sedimentary LABs around Brunei Bay with those from South East Asia and around the world suggests that sewage contamination in Brunei can still be considered low (Table 3).

The current study showed that the first station (SBB1) has the highest level of LABs compared to other stations along Brunei Bay. Higher urbanization and industrialization along Brunei Bay are likely to be responsible for these spatial distributions. The sedimentary LAB concentrations in Brunei Bay were heterogeneously distributed depending on the intensity of LAB inputs as well as TOC content of the sediments (Wang et al., 2001), indicating that the geographical distance of sampling stations from input sources do impact LAB distribution. These results are different from those of the studies of Takada and Ishiwatari (1991) and Raymundo and Preston (1992) suggesting that LAB concentrations throughout Tokyo Bay and Humber-Wash region were distributed uniformly.

The highest concentration of LABs is reported at SBB1 (41.3 ng g^{-1} dw), which is situated the farthest from Brunei Bay adjacent

Table 2
Linear alkylbenzenes (LABs) composition, concentration and relative compound ratio in the sedimentary surface from Brunei Bay (SBB1–SBB14).

	SBB1[a]	SBB2	SBB3	SBB4	SBB5	SBB6	SBB7	SBB8	SBB9	SBB10	SBB11	SBB12	SBB13	SBB14
∑ C10-LABs[b]	2.1	3.2	2.0	1.8	1.7	1.5	2.1	1.6	1.1	1.9	1.3	1.2	1.2	1.8
∑ C11-LABs	8.2	8.0	8.0	2.2	3.7	2.1	5.1	2.0	2.2	2.8	3.3	1.4	1.7	2.2
∑ C12-LABs	12.2	9.3	9.8	1.8	4.3	1.5	6.6	1.6	2.2	3.2	4.6	1.2	1.4	2.2
∑ C13-LABs	15.2	14.6	16.3	2.2	7.5	1.7	10.3	2.0	1.5	8.6	11.3	1.4	1.7	2.3
∑ C14-LABs	3.6	5.8	4.3	2.0	3.0	1.9	3.0	1.8	2.1	3.7	4.1	2.0	1.4	2.2
∑ LABs	41.3	40.9	40.4	10.1	20.2	8.6	27.1	9.0	9.2	20.2	24.6	7.1	7.4	10.8
LC-LABs (ng g^{-1} dw)[c]	18.8	20.4	20.6	4.3	10.5	3.6	13.3	3.8	3.6	12.3	15.3	3.3	3.1	4.6
SC-LABs (ng g^{-1} dw)[d]	10.3	11.2	10.0	4.0	5.4	3.5	7.2	3.6	3.4	4.7	4.6	2.6	2.9	4.0
I/E[e]	1.9	1.8	1.7	0.6	2.2	1.1	2.0	2.1	0.6	2.1	1.8	1.0	2.0	1.6
LAB degradation (%)[f]	38	35	34	−4.77	42	17	39	40	−5.4	41	36	16	40	31
TOC (%)[g]	1.6	1.6	0.4	0.1	1.0	0.1	1.5	0.9	1.3	1.9	0.4	0.3	0.6	0.7
TOC (mg/g)	15.6	16.2	3.8	1.1	10.0	1.4	14.6	9.1	12.5	19.2	3.5	2.7	5.5	6.6
∑ LABs/TOC (ng/mg)[h]	2.7	2.5	10.6	9.5	2.0	6.1	1.9	1.0	0.7	1.1	7.0	2.6	1.3	1.6

[a] SBB1, the first letter indicates the station, the second and third letters represent the first letters of location name, the numbers 1, 2, 3, 4, and 5 indicate the first, second, third, fourth and fifth stations for each location respectively.
[b] ∑ C10-LABs, sum of the 26 LAB congeners ranging from 5-C_{10} to 2-C_{14}.
[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-C_{11}.
[e] I/E (C12-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).
[f] LAB degradation (%), LAB deg = 81 × log (I/E ratio) + 15 (r^2 = 0.96).
[g] TOC (%), total organic carbon.
[h] ∑ LABs/TOC, ratio of total LABs over TOC.

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Table 3
The concentrations of LABs and I/E ratios around the world.

Location	N	Maximum ∑LABs[a](ng g^{-1})	I/E ratio[b]	Degradation[d] (%)	Reference
Sumidagawa River, Japan	6	12,110	1.7	34	Takada and Ishiwatari (1987)
Tamagawa River, Japan	8	15,790	1.9	38	Takada and Ishiwatari (1987)
Arakawa, Japan	1	4010	1.2	21	Takada and Ishiwatari (1987)
Nakagawa, Japan	1	2350	1.7	34	Takada and Ishiwatari (1987)
Edogawa, Japan	1	910	1.8	36	Takada and Ishiwatari (1987)
Yorogawa, Japan	1	120	1.4	27	Takada and Ishiwatari (1987)
Tokyo, Japan	1	590	1.5	29	Takada and Ishiwatari (1987)
Humber Estuary and Wash, UK	18	84.8	2.1	41	Raymundo and Preston (1992)
Thames, UK	5	2300	3.1	55	Raymundo and Preston (1992)
Tokyo Bay, Japan	29	3270	2.5	47	Takada et al. (1992)
Tokyo Bay, Japan	24	2750	3.1	55	Takada et al. (1992)
Tokyo Bay, Japan	44	2.45	6.3	80	Chalaux et al. (1995)
Victoria Harbor,China	12	23,500	2.6	49	Hong et al. (1995)
Southern California Bight, USA	67	92.9	4.6	69	Macías-Zamora and Ramírez-Alvarez (2004)
Japan	19	5860	6	78	Isobe et al. (2004)
Thailand	36	14,100	5.9	77	Isobe et al. (2004)
Malaysia	13	8590	4.8	70	Isobe et al. (2004)
Philippines	15	13,000	2.9	55	Isobe et al. (2004)
Cambodia	11	4200	1.7	34	Isobe et al. (2004)
Vietnam	35	8650	2.2	49	Isobe et al. (2004)
Indonesia	20	42,600	2.1	41	Isobe et al. (2004)
India	12	4450	2.1	41	Isobe et al. (2004)
Brunei Bay, Brunei	14	41	1.9	38	Present study
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	25.8	1.5	29	Luo et al. (2008)
South China Sea	28	23.1	0.9	11	Luo et al. (2008)
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)
Chaohu Lake	61	5270	2.1	41	Wang et al. (2012)
Sarawak River, Malaysia	9	7386.2	0.98	14	Magam et al. (2012)
Sembulan River,Malaysia	6	5567.1	1.8	36	Magam et al. (2012)
Dongjiang River	45	410	1.4	27	Zhang et al. (2012)
Outfalls of paper mills	3	3270	1.3	24	Zhang et al. (2012)
Admiralty Bay, Brazil	4	46.5	0.9	11	Martins et al. (2012)
Jakarta Bay	7	86,745	0.92	12	Rinawati et al. (2012)
Tokyo Bay	2	1109	2.8	51	Rinawati et al. (2012)
Jakarta city	13	1,559,373	1.3	24	Rinawati and Takada (2013)
Coastal zone off South China	30	160	1.2	21	Liu et al. (2013)
Babitonga Bay, Brazil	19	413.3	2.9	55	Martins et al. (2014)
Coastal shelf of China	32	77	1.2	21	Wei et al. (2014)
Detergents	10	5,303,000	1.7	34	Raymundo and preston (1992)
Detergents	12	702,000	1.1	18	Takada and Ishiwatari (1987)
Detergent		5,040,000			Eganhouse et al. (1983)
Liquid detergents			1.2	21	Ni et al. (2008)
Powder detergents			1.1	18	Ni et al. (2008)
Shampoos			1.6	32	Ni et al. (2008)

N = the number of samples.
[a] ΣLAB = sum of concentration of all secondary LAB congeners having C10–C14 alkyl chains.
[b] I/E = (6-C_{12} + 5-C_{12}) / (4-C_{12} + 3-C_{12} + 2-C_{12}).
[d] LAB deg = 81 ∗ log(I/E ratio) + 15(r^2 = 0.96).

to the open South China Sea. This is consistent with the findings of Wei et al. (2014) and Luo et al. (2008) where high concentrations of hydrophobic organic chemicals in the area were reported. Furthermore, high concentrations of LABs at the station could be attributed to a lateral transport of LABs from the poorly treated or untreated domestic sewage by the river to this station. Detailed studies should be conducted to confirm this suggestion. Brunei Bay has experienced rapid population growth in recent years and the detection of LABs which implied inputs of poorly treated and untreated domestic sewage into the bay should be of concern. LABs may have originated from hot spot areas such as hospitals, hotels, laundries, schools, governmental institutions, shopping centers, parks and restaurants where human activities are intense. Another factor that contributed to the elevated levels of LABs in SBB1 is high concentration of TOC at 15.6 mg/g dw. A positive correlation between the concentrations of hydrophobic organic compounds (HOCs) and TOC was previously reported by several workers (Arzayus et al., 2001; Accardi-Dey and Gschwend, 2002; Hinga, 2003).

The lowest concentration of LABs was found at station SBB12 of Brunei Bay (7.1 ng g^{-1} dw). This can be explained by less anthropogenic influence around the station. In general, the results of this study suggested that Brunei Bay is subjected to low impact from domestic wastewater discharge.

Prior studies have documented that wastewater discharge combined with horizontal flow are the two main culprits for the occurrence of LABs in the Bay sediments (Sherblom et al., 1992; Luo et al., 2008; Ni et al., 2009). Atmospheric movement and hydrodynamic flow were input routes for hydrophobic contaminants in soils (Wania et al., 1998; Jones and de Voogt, 1999). Brunei Bay is teeming with fishing activities that directly discharge sewage into the marine environment. This explains the higher levels of ∑LABs in the SBB1, SBB2 and SBB3 (41.3, 40.9 and 40.4 ng g^{-1} dw respectively). At the same time, due to the greater consumption of alkyl benzene sulfonate-type detergents around these stations, the amount of lateral transport of LABs to the coastal area is higher. Additionally, LABs in the coastal areas are better

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preserved due to water depths. Lower levels of SBB4 (10.1 ng g^{-1} dw) may be attributed to less intensive coastal fishing and tourism activities at this station than at other stations. SBB4 is located at the mouth of Brunei Bay, where frequent intense rainfall supplies large amounts of eroded soil into the river; thus, dilution at SBB4 could be more efficient than at other locations. The relatively lower LAB concentrations may also relate to the solid settling stage during the primary treatment. This stage can remove some of the LABs before discharge in the bay (Eganhouse and Sherblom, 2001). In a similar way, since SBB6, SBB8, SBB9, SBB12, SBB13 and SBB14 are less urbanized, there was a lower concentration of LABs in this area. The distribution of LABs in the marine environment is controlled by lateral transport of sewage particles and dilution by natural particles in addition to the intensity of sewage inputs (Zeng et al., 1997; Isobe et al., 2004). Eganhouse et al. (1983) and Valls et al. (1989) observed a decrease in LAB concentration and depletion of external LABs rather than internal ones in offshore sediments compared to inshore or estuary sediments.

Table 2 shows that the isomeric compositions and the chain length distributions of LABs in sediments were relatively abundant with C_{13}, C_{12} homologs, respectively. C_{13} homologs were the most abundant and found in most of the stations at Brunei Bay (Table 2), while C_{10} and C_{14} homologs were found in much lower abundance in most of the stations. This is consistent with Phillips et al. (2001) who reported that LABs in bottom sediments at the near-outfall and mid-shelf sites of southern California were enriched with C_{13} and C_{12} homologs, while the proportions of C_{10} and C_{14} homologs were found to be lower. These differences likely reflect selective losses of the more soluble C_{10} and C_{14} homologs during settling of effluent particles. On the other hand, the isomeric compositions as shown in Table 2 reveal that the chain length distributions of LABs in sediments with C_{10}, C_{11} homologs decreased in abundance compared to those in detergent and sewage sludge (Luo et al., 2008). The relative abundance of the C_{13} homolog group was obviously apparent in all sediment samples, which is consistent with previous studies (Luo et al., 2008). More rapid or selective degradation of short homolog groups (C_{10}, C_{14}) in the environment could be more probable. Therefore, the ratio of C_{13}/C_{12} was calculated for all sediments. As household detergents are the major source of LABs in the environment, the population may be a major factor in the distribution of LABs in the aquatic environment (Ni et al., 2008).

A good positive correlation between LABs and organic matter content is expected because of the hydrophobicity of LAB molecules. Therefore, the TOC is frequently used as a normalizer (Krumgalz, 1989; Grathwohl, 1990). Tropical rainfall results in abundant plant life, therefore, there are high amounts of organic matter in Malaysia (Vaezzadeh et al., 2015). The spatial variability of $\sum$LAB concentrations in sediments can be a result of both environmental parameters (i.e., TOC content and input intensities). Fig. 2. shows that there is linear relationship between TOC and $\sum$LABs at Brunei Bay ($R^2 = 0.5$; Fig. 2) at $p < 0.05$. This could suggest that sediment TOC is a decisive factor in distribution of LABs in the surface sediments of Brunei Bay. Hydrophobic compounds such as LABs tend to partition into sediment from pore water due to organic carbon content of the sediment (McGroddy and Farrington, 1995). Therefore, the sedimentary TOC from domestic sewage can be the controlling factor in distribution of LABs, however, this linear relationship may also indicate that there are other controlling factors such as local sewage discharges giving rise to the levels of LABs and TOC in the sediments. This confirmed a previous hypothesis that a significant linear correlation between the concentrations of $\sum$LABs and TOC indicates wastewater discharge as the dominant input route for organic carbon into sediments of the river (Zhang et al., 2012). However, others reported a lack of correlation between hydrophobic compounds and TOC in sediment (Vaezzadeh et al., in press).

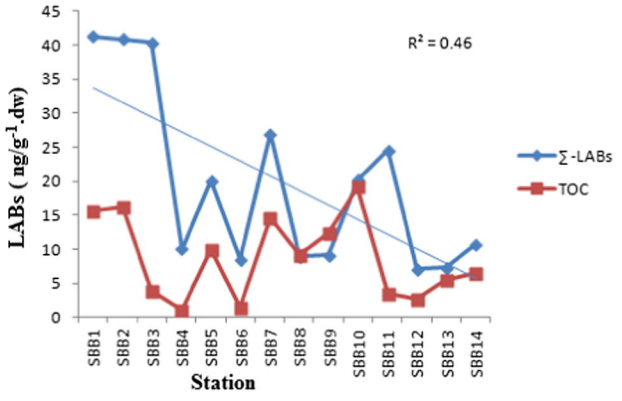

Fig. 2. Correlation between LAB concentration and TOC in Brunei Bay sediments.

The ratios of LABs have been reported to be useful indicators for determining their source (Takada and Eganhouse, 1998). The I/E ratio (6-C_{12}LAB + 5-C_{12}LAB / 4-C_{12}LAB + 3-C_{12}LAB + 2-C_{12}LAB) increases or systematically changes during LAB degradation under aerobic conditions (Takada and Ishiwatari, 1990), while the relative abundance of LAB isomers in the commercial detergents and in raw sewage are equal (Takada and Ishiwatari, 1987). Thus, this ratio has been proposed (Takada and Ishiwatari, 1990) and applied (Takada and Ishiwatari, 1990; Isobe et al., 2004; Macías-Zamora and Ramírez-Alvarez, 2004) as an indicator of the extent of LAB degradation. All sedimentary I/E ratios in all stations in the present study ranged from 0.56 to 2.2 as shown in Fig. 3. These results of the study indicated that primary and secondary effluents were discharged into riverine and coastal waters (Fig. 3). The sediment I/E ratios in Brunei Bay stations were slightly higher than those in the Pearl River Estuary (0.6–1.5) (Luo et al., 2008), which may suggest a relatively greater discharge of treated wastewater in Brunei Bay. Takada and Eganhouse (1998) suggested that the I/E ratio related properly to the extent of LAB degradation and the relationship is derived using log linear regression analysis estimated to be: LAB deg (%) = 81 × log (I/E ratio) + 15 (r^2) = 0:96. LAB degradation values were calculated for all surface sediment samples of Brunei Bay. Using I/E LAB C_{12}, the values of LAB degradation in Brunei Bay ranged from 0–42% (Table 2). The lower degradation of LABs in Brunei sediments (SBB4 and SBB9) can be explained by the fact that the LAB compounds are located within anaerobic conditions and this has caused limited degradation of LABs. On the other hand the higher LAB degradation in sediments of the rest of the stations of Brunei Bay emphasized that these LABs are located in an aerobic environment and most of them have degraded in the water column and via recent inputs deposited on the water bottom. High LAB degradation indexes in Brunei Bay indicated an improvement in the wastewater treatment, thus reducing its recent discharges. Compared with near shore sediments from the PRE and the adjacent South China Sea (Luo et al., 2008), a relatively higher abundance of C_{13}-LABs in Brunei sediments possibly points to a greater extent of degradation as it is located farther from inland sources.

Conclusion

Generally, the LAB concentrations in surface sediments of Brunei Bay were low as compared to other South and South East Asian countries. The highest and lowest LAB concentrations in Brunei Bay were detected in the first station and the twelfth station, respectively. The I/E ratios were found to be high, demonstrating the presence of primary and secondary sewage effluent sources into Brunei Bay. There is no previous data in Brunei Bay that could be used to evaluate the trend in the LAB distribution and concentrations. LABs are powerful tools to trace sewage contamination. Since Brunei Bay is situated in an ecologically sensitive area and intensively used for fishing activities, the Bay ecosystem requires continuous monitoring for contamination due to sewage pollution in the near future.

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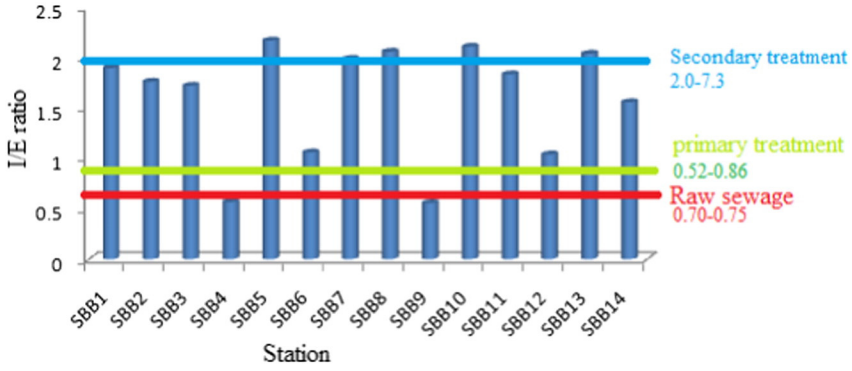

Fig. 3. The I/E ratio in Brunei Bay samples (the horizontal lines are stated as in Takada and Eganhouse, 1998).

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