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Mercury contamination in the estuaries and coastal sediments of the Strait of Malacca

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Abstract Sediment is a great indicator for assessing coastal mercury contamination. This work profiled the magnitude of mercury pollution in the tropical estuaries and coastal sediments of the Strait of Malacca. Mercury was extracted through the ultrasound-assisted mercury extraction method and analyzed using the flow injection mercury system. The mean concentration of mercury in the sediment samples was 61.43±23.25 μg/kg, ranging from 16.55±0.61 to 114.02±1.54 μg/kg. Geoaccumulation index revealed that a total of 13 % of sampling sites were moderately enriched with mercury. The northern part of the Strait of Malacca had the highest mean mercury (Hg) concentration (76.36± 27.25 μg/kg), followed by the southern (64.59± 16.09 μg/kg) and central (39.33±12.91 μg/kg) parts. Sediment mercury concentration in the current study was lower than other regions like Japan, China, Indian, east Mediterranean, and Taiwan. When compared to the Canadian interim marine and freshwater sediment, China's soil interim environmental guidelines, mercury contamination in the Strait of Malacca was found to be below these permissible limits. Sediment organic matter content was found to have significant correlation with sediment mercury concentration. This study could provide the latest benchmark of mercury pollution and prove beneficial to future pollution studies in relation to monitoring works in tropical estuaries and coastal sediments.

Keywords Estuaries · Coastal sediment · Geoaccumulation index · Mercury analyzer · Mercury contamination · Strait of Malacca

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Introduction

Since the outbreaks of the Minamata disease in the 1950s/60s at Minamata Bay, Japan, a great deal of attention has been paid to mercury contamination (Yoshida 2012). Mercury is a highly toxic pollutant that is cycled between the environmental compartments (i.e., air, water, sediment, and biota) (Canário et al. 2005). Practically, three major chemical forms of mercury exist in the environment: elemental or metallic mercury (Hg^0), inorganic mercury (Hg^{2+}), and organic mercury (CH_3Hg^+) (Cornelis et al. 2005). Among all the forms of

mercury species, the organic mercury compound, particularly methylmercury, is the most toxic form of mercury present in the environment (Gochfeld 2003; Zahir et al. 2005). Owing to its lipid solubility, organic mercury has the ability to bioaccumulate and biomagnify within the food chain once released into the environment (Zahir et al. 2005; Maggie et al. 2009). It can easily cross the blood-brain barrier and demonstrate adverse health effects on human dietary exposure to mercury (Zahir et al. 2005; Hafshejani et al. 2012). Elevated mercury concentration in the human body can lead to severe impairment, such as neurological, nephrological, immunological, cardiac, motor, reproductive, and genetic disorders (Gochfeld 2003; Cornelis et al. 2005; Zahir et al. 2005).

Mercury is released into the environment from both natural and man-made sources (Haris and Aris 2012). The contribution of volcanic eruptions, geothermal sources, and re-emissions of historically deposited mercury in the sediment and sea surfaces are natural sources of mercury emitted into the environment (Pirrone et al. 2010). In addition, mercury is also released to the environment via anthropogenic sources, such as the burning of fossil fuels, waste incineration, paint manufacturing, industrial activities, coal-fired power plants, mining and smelting, usage of pesticides, and pulp and paper manufacturing (F rstner and Wittman 1979; Ainza et al. 2010; Pirrone et al. 2010; Haris and Aris 2012; Wu et al. 2013). When effluents containing mercuric species are discharged into rivers, they tend to stick to particulates, especially organic matter in the water (Ram et al. 2009). During the tidal mixing event, mercury bound particles tend to settle out with water and making sediments as a great sink of mercury in aquatic environment before being transported and bioaccumulated within a trophic level (Ram et al. 2009; Praveena et al. 2008, 2010; Haris and Aris 2012; Chen et al. 2013).

Tropical estuaries and coastal systems play an important role in the biogeochemical cycling of mercury in the environment (Fig. 1.). According to Smoak and Patchineelam (1999), tropical coastal systems contribute approximately 75 % of sediment discharge from inland to the sea (Senthilkumar et al. 2013). They provide great sinks for mercury since they have the ability to retain contaminants from various pollution sources, such as tidal waters, riverine input, and surface water runoff (Senthilkumar et al. 2013; Tam and Wong 1997). Therefore, the distribution and degree of contamination of mercury in the sediment can serve as a pollution indicator in estuaries and coastal sediments (Yap et al. 2003a; Ram et al. 2009).

Since there are only limited studies regarding mercury concentration and its distribution in tropical surface sediments (Law and Singh 1987; Kannan and Falandysz 1998; Yap et al. 2003b; Hajeb et al. 2012; Haris and Aris 2012), a comprehensive study working on mercury concentration along the Strait of Malacca is urgently required. The objective of the present study is to provide

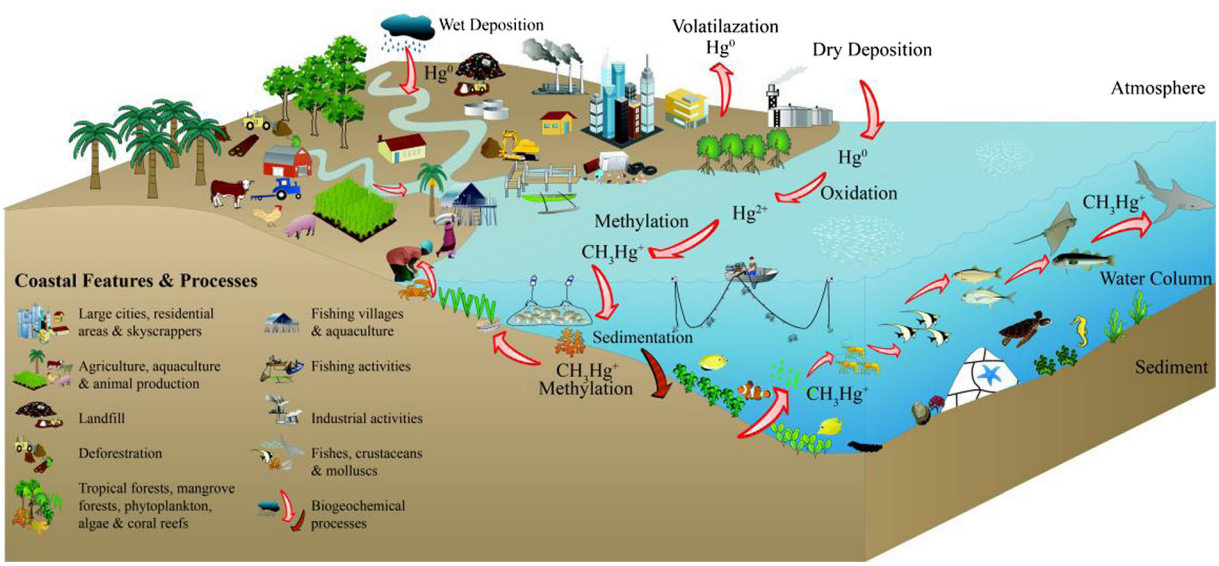

Fig. 1 Biogeochemical cycling of mercury in tropical estuaries and coastal system (symbols for diagrams courtesy of the Integration and Application Network, University of Maryland Center for Environmental Science, ian.umces.edu/symbols)

the latest information on the mercury concentration and pollution in surface estuaries and coastal sediments along the Strait of Malacca. In addition, the mercury concentrations detected in this study were compared with the Canadian interim marine and freshwater sediment guideline for mercury (Gaudet et al. 1995) and interim environmental guidelines for mercury in soil in China (Wu et al. 1991). Ultimately, the geoaccumulation index (I_{geo}) was used to determine the degree of mercury contamination of the estuary and coastal sediment along the Strait of Malacca.

Methods

Sampling sites

The west coast of Peninsular Malaysia is concentrated with industrial, agricultural, and economic activities owing to the availability of abundant natural resources for use by humans (Abdullah et al. 1999; Shazili et al. 2006). The current study covers the coastal areas and estuaries of the west coast of Peninsular Malaysia from north Kuala Kedah to South Tanjung Kupang along the Strait of Malacca (Fig. 2.). During sampling, water physico-chemical parameters (pH, electrical conductivity (EC), and salinity) were measured in situ. In addition, surface coastal and estuaries samples were collected from 56 sampling stations during the month of February to May 2012. Table 1 summarizes the general site descriptions and number of stations for each sampling point. The sampling locations were identified prior to the sampling activities in such a way that they could be accessible and precisely represent the current mercury pollution condition and pollution sources.

Samples collection and analytical procedure

Homogenized surface sediment (0–5 cm) samples were collected at low tide from the sampling sites using hand-held plastic sterile scoops. The samples were then transferred to acid-washed double-ziplock polyethylene bags and stored in an icebox (<4 °C) prior to transporting to the laboratory. In the laboratory, sediment samples were naturally air-dried at room temperature until constant weight was achieved before they could be subjected to further laboratory analyses. Subsequently, the sediment samples were agitated by using pestles and mortars and sieved to obtained particle sizes of <2 mm for physico-chemical parameters analyses and <63 μm for mercury analysis.

The physico-chemical parameters (pH, electrical conductivity (EC), salinity (Sal)) of the water and sediment samples were measured using WTW pH330i (WTW Wissenschaftlich-Technische, Germany) and YSI 63 (YSI Inc., Yellow Springs, Ohio). For the determination of the physico-chemical characteristics in the sediment samples, the sediment to Milli-Q® water ratio of 1:2 was prepared in a beaker and allowed to stand for 30 min. The mixture was stirred every 10 min during the suspension. Then, it was let stand for an hour before the measurements were taken in triplicate (Radojević and Bashkin 2006). On the other hand, approximate total organic carbon of sediment samples were determined by loss on ignition (LOI) method (Radojević and Bashkin 2006). Approximately 2 g of sediment samples (<2 mm) were weighed into a crucible and oven-dried at 105 °C sediment for 12 h. After cooling the sediment samples to room temperature, the samples were weighed again. Next, the samples were placed in a preheated muffle furnace and ignited at 550±25 °C until constant weight was achieved. The cooled samples were weighed. The three weights measured were used to calculate percent of organic matter (%OM) as LOI. Next, sediment particle sizes were determined based on hydrometer test in ASTM D 422-63 method (ASTM 1963). The sediment samples were divided into sand (>50 μm), silt (2 μm< size<64 μm), and clay (<2 μm) fraction according to the USDA particle size classification (SSDS Soil survey division staff 1993).

In order to determine the mercury in the sediment samples, the mercury extraction procedure proposed by (Collasiol et al. 2004) was employed with some modifications. This extraction method was chosen owing to its accuracy and efficiency of mercury leaching over conventional digestion methods. Firstly, 1.0 g of the sieved sediment sample was weighed and placed into a 50-mL volumetric flask. Then, 15 mL of 30 % nitric acid, HNO_3 (Merck, Darmstad, Germany), was added into the volumetric flask. The mixture was allowed to stand for 30 min. Subsequently, 0.155 (*m*/*v*) of potassium chloride, KCl salt (Merck), was then added to enhance the mercury leaching from the sediment sample (Collasiol et al. 2004). The mixture was then subjected to the sonication process in a bath sonicator at 70 W for 180 s. An iced water bath was used during the sonication process to avoid mercury loss by volatilization due to heating. Later, the extracted sample solution was

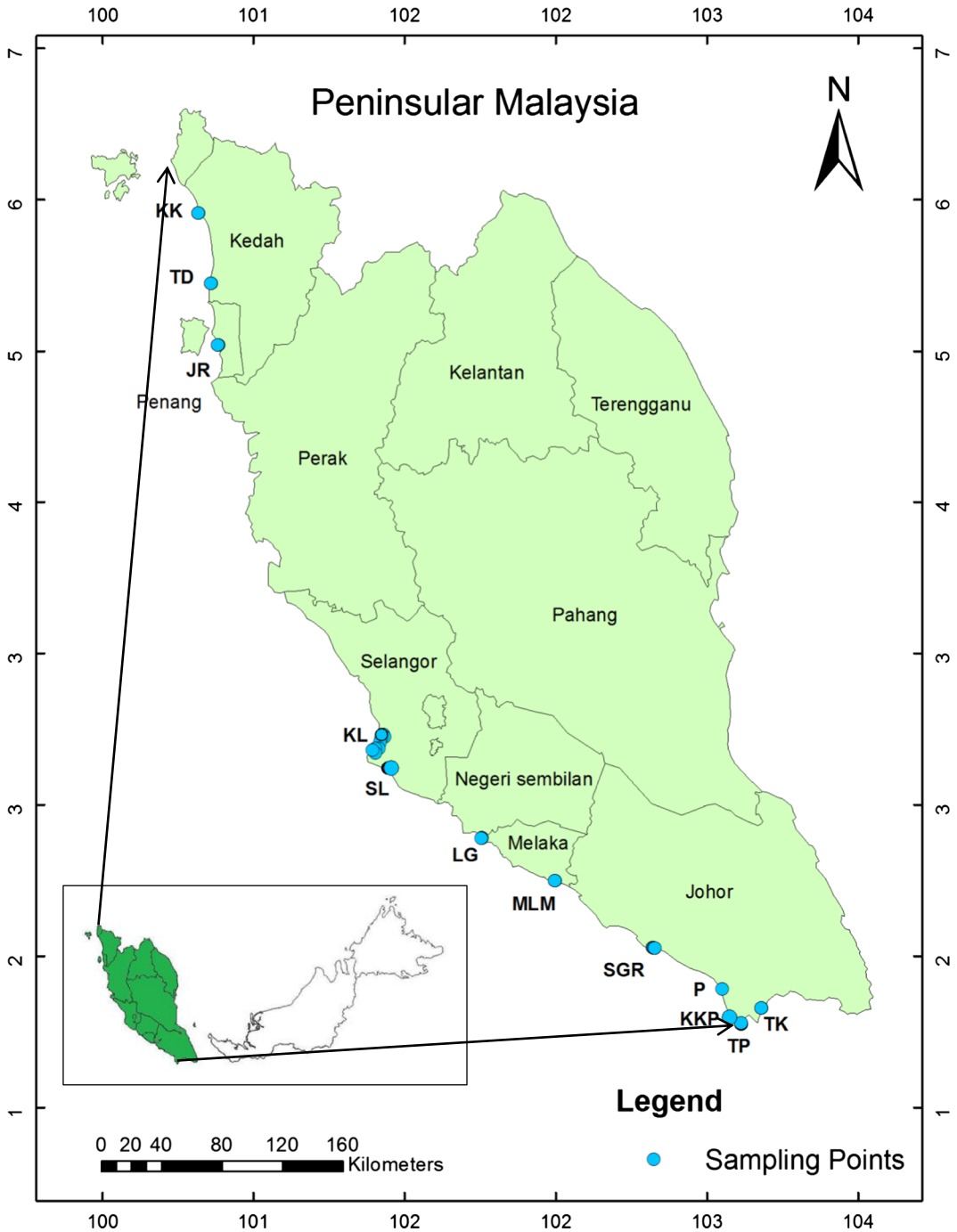

Fig. 2 Study area showing the sampling stations

centrifuged at 2700 rpm for 2 min to settle the particulates. Finally, the extracts were analyzed for mercury concentration by using the mercury analyzer flow injection mercury system (FIMS 400, PerkinElmer).

Quality assurance and quality control

Quality assurance and quality control of the field and analytical works are essential in order to

Table 1 Description of sampling sites

No.	Sites	Abbreviation	GPS	Site Description	State
1	Kuala Kedah	KK	N 6° 6.240' E 100°17.102'	Jetty in the vicinity, shipping and fishing activity	Kedah
2	Kuala Kedah		N 6° 6.217' E 100°17.113'	A dock, boat parking	
3	Kuala Kedah		N 6° 6.209' E 100°17.112'	Near to a restaurant	
4	Kuala Kedah		N 6° 6.199' E 100°17.111'	A fishing site	
5	Kuala Kedah		N 6° 6.253' E 100°17.093'	A site near to a mangrove area	
6	Tanjung Dawai	TD	N 5° 41.101' E 100°21.571'	A fishing site facing open sea	
7	Tanjung Dawai		N 5° 41.099' E 100°21.567'	A site near to residential area	
8	Tanjung Dawai		N 5° 41.102' E 100°21.562'	A site near to a mangrove area	
9	Tanjung Dawai		N 5° 41.106' E 100°21.558'	A fishing site	
10	Tanjung Dawai		N 5° 41.113' E 100°21.554'	A fishing site	
11	Kuala Juru	JR	N 5° 20.348' E 100° 24.232'	Near to quarry mining	Penang
12	Kuala Juru		N 5° 24.411' E 100° 24.394'	Estuary with industrial activities in the vicinity	
13	Kuala Juru		N 5° 20.394' E 100° 24.461'	A side near to factory, industrial activities, opposite to fishing village	
14	Kuala Juru		N 5° 20.367' E 100° 24.391'	Fishing village in vicinity, a fishing site	
15	Kuala Juru		N 5° 20.297' E 100° 24.301'	River mouth to open sea	
16	North port Klang	KL	N 3° 0.093' E 101° 22.670'	Heavily shipping lane, industrial activities in the vicinity	Selangor
17	Klang River		N 3° 0.143' E 101° 23.272'	River mouth of Klang river, heavily shipping lane, bad odour	
18	South port Klang		N 2° 59.304' E 101° 23.640'	River mouth, industrial and shipping activities	
19	Lumut Strait		N 2° 58.715' E 101° 22.435'	A site near mangrove area between Pulau Indah and Pulau Tonggok	
20	Lumut Strait		N 2° 57.367' E 101° 22.003'	Estuary of Telok Gong, industrial activity in the vicinity	
21	Klang - Langat	KL	N 2° 55.289' E 101° 21.477'	River mouth of Langat river, mangrove area, shipping and fishing activities	Selangor
22	Klang - Langat		N 2° 55.259' E 101° 20.327'	A site at Pulau Carey, mangrove area, shipping activities	
23	Klang - Langat		N 2° 53.729' E 101° 20.191'	A site between Pulau Indah and Pulau Carey, shipping and fishing activities	
24	Klang - Langat		N 2° 54.648' E 101°19.208'	Busy shipping lane and mangrove area in the vicinity	
25	Klang - Langat		N 2° 53.204' E 101° 18.062'	Mangrove areas which is opposite to the dockyards	
26	Langat River	SL	N 2° 48.228' E 101° 24.531'	Downstream of river, a fishing site	
27	Langat River		N 2° 48.363' E 101° 24.668'	A site at river mouth	
28	North port Klang	K	N 3° 0.169' E 101° 22.451'	Jetty with boating activity	
29	North port Klang		N 3° 0.131' E 101° 22.423'	A site in mangrove area which near to a small jetty	
30	Senggarang	SGR	N 1° 44.330' E 102° 59.180'	Near to village	Johor
31	Senggarang		N 1° 44.337' E 102° 59.981'	Small river mouth	
32	Senggarang		N 1° 44.328' E 103° 0.094'	A fishing site	
33	Senggarang		N 1° 44.318' E 102° 59.910'	Near to palm oil plantation	
34	Senggarang		N 1° 44.299' E 103° 0.140'	Agricultural activity	
35	Kampung Perpat Pasir	TP	N 1° 17.044' E 103° 30.622'	A site near to a river, shipping activities	
36	Kampung Perpat Pasir		N 1° 17.056' E 103° 30.620'	A site near to busy road	
37	Kampung Perpat Pasir		N 1° 17.019' E 103° 30.625'	Heavy shipping lane and dockyards nearby	
38	Kampung Perpat Pasir		N 1° 17.204' E 103° 30.649'	A site near to mangrove area, fishing activity	
39	Kampung Perpat Pasir		N 1° 17.295' E 103° 30.653'	A site near to a small river and mangrove area	
40	Tanjung Kupang	TK	N 1° 22.721' E 103° 38.093'	Fishing village, boat parking	Johor
41	Tanjung Kupang		N 1° 22.673' E 103° 38.113'	Fishing village, near to small river	
42	Tanjung Kupang		N 1° 22.679' E 103° 38.113'	A site near to mangrove area	
43	Tanjung Kupang		N 1° 22.672' E 103° 38.114'	Estuary, boats	
44	Tanjung Kupang		N 1° 22.717' E 103° 38.094'	Fishing vilage near to food stalls	
45	Kukup	KKP	N 1° 19.403' E 103° 26.626'	Fishing village, near to jetty to Batam Island and Tanjung Balai	
46	Kukup		N 1° 19.348' E 103° 26.658'	Fishing village, neat to jetty to Batam Island and Tanjung Balai	
47	Kukup		N 1° 19.315' E 103° 26.689'	Mangrove areas in the vicinity	
48	Pontain	P	N 1° 29.378' E 103° 23.128'	Small town in the vicinity, near to river mouth of Sg. Pontain Kechil	
49	Kuala Merlimau	MLM	N 2° 7.971' E 102° 24.236'	A small jetty, wet market	Melaka
50	Kuala Merlimau		N 2° 7.953' E 102° 24.230'	A site with construction activity, a lot of shells	
51	Kuala Merlimau		N 2° 7.941' E 102° 24.223'	Estuary near to mangrove area	
52	Kuala Merlimau		N 2° 7.929' E 102° 24.213'	A site at river mouth	
53	Kuala Merlimau		N 2° 7.928' E 102° 24.183'	A site along the shore	
54	Kuala Linggi	LG	N 2° 23.370' E 101° 58.324'	A site with fishing and shipping activities	Negeri Sembilan
55	Kuala Linggi		N 2° 23.313' E 101° 58.292'	A site near to restaurant	
56	Kuala Linggi		N 2° 23.259' E 101° 58.155'	A site near to recreational activity	

ensure that highly precise and representative mercury concentrations of sediment are produced (Praveena and Aris 2012). Triplicate and homogenized sediment samples were collected to estimate the variability resulting from the sampling activities. Each probe used in this study was calibrated with a calibration solution prior to measurement. Triplicate measurements (n=3) were taken and tested with relative standard deviation to ensure highly reliable and accurate results (APHA 2005). The reagents used in the current analysis were all of analytical reagent grade. Ultrapure water (water sensitivity >18.2 Mohms·cm at 25 °C; Millipore, MA, USA) from Milli-Q® system was used for laboratory applications including reagent and blank preparation and standard preparation as well as equipment and apparatus rinsing activity. In addition, the laboratory glassware used was acid-washed with 10 % (v/v) HNO_3 for 24 h and the flasks to contain extracted samples were soaked with 33 % (v/v) HNO_3 for a week before they were then rinsed with ultrapure water. Moreover, equipment and sample blanks were used to ascertain the background correction. A series of standard solutions were prepared by diluting PerkinElmer Pure mercury stock solution (PerkinElmer, Connecticut, USA). Recovery tests were performed to check the accuracy of data before and after each day of measurements. In addition, the accuracy of the analytical procedure was checked with standard reference material for estuarine sediment (National Institute of Standards and Technology, estuarine sediment 1464a, Gloucester Point, VA). The mercury recovery was satisfactory (±10 %), ranging from 97 to 108 %. In addition, the sensitivity of the method was determined by calculating the limit of detection (LOD) and limit of quantification (LOQ). According to Sanagi et al. (2009), the LOD and LOQ are the concentrations of the analyte that would yield signal-to-noise ratios of 3 and 10, respectively. The LOD represents the lowest concentration of analyte that is detectable and reliably distinguished from zero, but cannot necessarily be quantified, whereas the LOQ is the lowest concentration of analyte that can be determined quantitatively with an acceptable level of precision (González and Herrador 2007). In this study, the LOD and LOQ were calculated from ten blank samples using the following formula (González and Herrador 2007):

$$\mathrm{LOD} = \frac{3\mathrm{SD}_{\mathrm{blank}}}{b} \quad (1)$$

$$\mathrm{LOQ} = \frac{10\mathrm{SD}_{\mathrm{blank}}}{b} \quad (2)$$

where

$\mathrm{SD}_{\mathrm{blank}}$ Standard deviation of blank samples
b Slope of calibration curve

The LOD and LOQ calculated were 0.01 and 0.03 μg/kg, respectively. Therefore, the present analytical method for determination of the mercury concentration in sediments is sensitive and comparable to previous study by Collasiol and co-workers, which the LOD was detected to be 0.2 μg/l (Collasiol et al. 2004).

Table 2 Geoaccumulation Index (I_{geo}) contamination classification

I_{geo} value	Class	Designation of sediment quality
$I_{geo} \leq 0$	0	Practically uncontaminated
$0 < I_{geo} < 1$	1	Uncontaminated to moderately contaminated
$1 < I_{geo} < 2$	2	Moderately contaminated
$2 < I_{geo} < 3$	3	Moderately to strongly contaminated
$3 < I_{geo} < 4$	4	Strongly contaminated
$4 < I_{geo} < 5$	5	Strongly to extremely contaminated
$I_{geo} \geq 5$	6	Extremely contaminated

Calculation of mercury concentrations and geoaccumulation index

The concentration of mercury (Hg) in the sediment sample was calculated using the following equation (Haris and Aris 2012):

$$\text{Concentration in sediment (mg/kg)} = C \times \frac{V}{M} \quad (3)$$

where

C The concentration of mercury in the extract (in μg/mL)
V The volume of extract (in mL)
M The weight of sample (in g)

Table 3 Descriptive analyses of selected parameters for water and sediment of the Strait of Malacca

Matrix		Unit	Minimum	Maximum	Mean	SD	CV (%)
Sediment	pH	–	2.85	7.97	6.24	1.63	26.14
	EC	mS/cm	8.92	46.37	18.53	6.4	34.51
	Sal	ppt	4.97	30.13	11.04	4.24	38.4
	LOI	%	2.34	14.76	7.71	3.38	43.84
	Sand	%	1.05	70.04	14.20	17.11	120.51
	Silt	%	21.99	80.46	55.00	14.63	26.60
	Clay	%	4.40	62.00	30.76	15.42	50.13
	Hg	μg/kg	16.55	114.02	61.43	23.25	37.85
Water	pH	–	6.32	8.44	7.75	0.53	6.89
	EC	mS/cm	15.43	49.03	35.73	9.99	27.97
	Sal	ppt	7.43	32.20	21.92	7.33	33.44

SD standard deviation, *CV* coefficient of variation

In order to assess the degree of mercury pollution in estuaries and coastal sediments along the Strait of Malacca, the geoaccumulation index (I_{geo}) was calculated using Eq. 4, which was introduced by Müller (1969).

$$I\text{geo} = \log_2 \frac{C_n}{1.5 \cdot B_n} \quad (4)$$

where

C_n Mercury concentration in sediment analyzed (in μg/kg)

B_n Background concentration of mercury (in μg/kg)= 30 μg/kg

B_n is the average mercury concentration for sediment from the west coast of Peninsular Malaysia (Yap et al. 2003a, b).

This index is capable of being an effective and powerful method to elucidate the sediment quality of each sampling station (Krupadam et al. 2006; Praveena et al. 2007, 2008; Yap and Pang 2011; Haris and Aris 2012; Nillin et al. 2013). It calculates the estuary and coastal contamination by comparing the analyzed mercury concentration with its background concentration. In Eq. 4, the constant value of 1.5 is multiplied by the background mercury concentration in order to include the natural fluctuation of the mercury in the environment. In

Table 4 Correlation coefficient (*r*) of selected physico-chemical parameters and total mercury for water and sediment of the Strait of Malacca

	pH (w)	EC (w)	Salinity (w)	Hg (s)	pH (s)	EC (s)	Salinity (s)	LOI	Sand	Silt	Clay
pH (w)	1	0.265	0.214	0.097	0.005	0.367	0.360	0.095	−0.151	−0.131	0.310
EC (w)	0.001**	1	*0.983*	−0.361	0.090	0.037	0.040	−0.042	−0.268	0.012	0.301
Salinity (w)	0.010*	0.000**	1	−0.378	0.050	−0.055	−0.053	−0.036	−0.203	0.031	0.205
Hg (s)	0.249	0.000**	0.000**	1	−0.079	0.179	0.187	*0.510*	0.046	0.035	−0.090
pH (s)	0.955	0.283	0.552	0.348	1	0.020	0.028	−0.461	0.277	−0.344	0.029
EC (s)	0.000**	0.656	0.509	0.032*	0.809	1	*0.999*	0.227	−0.114	−0.239	0.378
Salinity (s)	0.000**	0.636	0.528	0.025*	0.736	0.000**	1	0.233	−.109	−0.236	0.370
LOI	0.259	0.619	0.667	0.000**	0.000**	0.006**	0.005**	1	−0.303	0.317	0.028
Sand	0.071	0.001**	0.015*	0.587	0.001**	0.173	0.192	0.000**	1	−0.59	−0.561
Silt	0.118	0.886	0.709	0.674	0.000**	0.004**	0.004**	0.000**	0.000**	1	−0.336
Clay	0.000**	0.000**	0.014*	0.286	0.729	0.000**	0.000**	0.741	0.000**	0.000**	1

The italic values are correlation values above 0.50 that were taken. Correlation value (upper triangle)

Significant value (lower triangle; **p value <0.01; *p value <0.05; two-tailed)

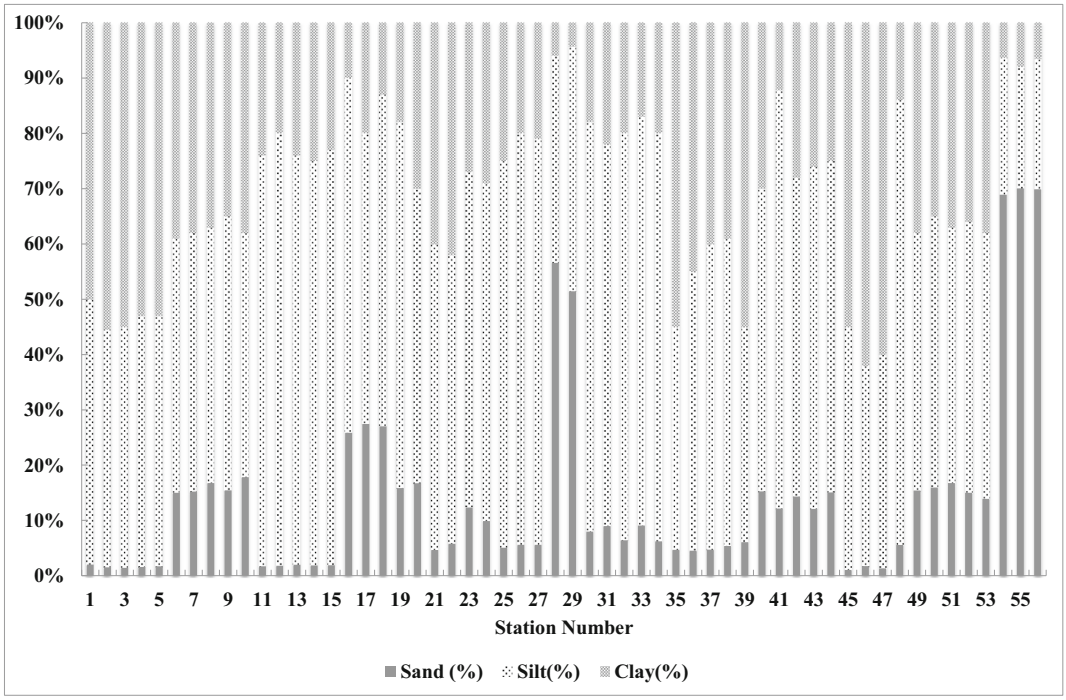

Fig. 3 Particle size distribution for sediments collected from 56 sampling stations along the Strait of Malacca

essence, the I_{geo} results can be classified into seven classes (Table 2).

Statistical analyses

In the present study, all the statistical analyses were performed using PASW Statistics 18. In order to avoid un-conservative conclusions, non-normally distributed raw data were not normalized prior to various statistical analyses (Lim et al. 2013). Descriptive statistics of selected physico-chemical parameters and Hg concentrations in sediments, such as mean, minimum, maximum, standard deviation, and coefficient of variance, were calculated. In addition, one-way analysis of variance (ANOVA) was conducted to measure the significant differences between the variables and sampling stations. Furthermore, Pearson product moment correlation analysis was performed in order to investigate the linear relationships between the measured variables. The value of correlation relationship (r) ranged from −1 to +1, where +1 represents a strong perfect positive relationship, 0 indicates no relationship, and −1 represents an absolute negative relationship between the variables. Additionally, for the ease of comparison purposes of the mercury, the sampling stations of estuaries and coastal sediments along the Strait of Malacca were divided into northern (stations 1–15), central (stations 16–29), and southern (stations 30–56) parts according to the GPS coordinates of the part. Then, the means of the Hg concentrations of the parts were statistically compared using Tukey-Kramer multiple comparisons at a confidence level of 95 %.

Results and discussion

Water and sediment characteristics

The descriptive statistics for selected physico-chemical parameters in the water and sediment are presented in Table 3. The one-way ANOVA shows there were significant differences ($p<0.05$; ANOVA) between the selected physico-chemical parameters. The coefficient of variances (CV) for all measured water and sediment parameters were greater than 30 %, an indication of a high variation in the water and sediment data along the Strait of Malacca with the exception of the pH in the water samples. High variations of these parameters might be attributed to the different water and sediment characteristics between the sampling stations. For instance, the sampling stations that received the influence

Table 5 Descriptive analyses of selected parameters according to regions for water and sediment of the Strait of Malacca

Region	Matrix		Unit	Minimum	Maximum	Mean	SD	CV (%)
Northern	Sediment	pH	–	4.32	7.97	6.69	1.39	20.81
		EC	mS/cm	13.78	46.37	23.40	8.16	34.86
		Sal	ppt	7.93	30.13	14.26	5.54	38.84
		LOI	%	3.65	14.01	8.26	3.43	41.55
		Sand	%	1.41	17.81	6.50	7.01	107.79
		Silt	%	43.09	78.22	55.54	14.35	25.83
		Clay	%	20.00	55.40	37.96	12.84	33.81
		Hg	μg/kg	29.89	114.02	76.36	27.25	35.68
	Water	pH	–	7.56	8.13	7.95	0.15	1.89
		EC	mS/cm	15.43	46.12	29.07	10.38	35.72
		Sal	ppt	7.43	26.93	15.78	6.72	42.61
Central	Sediment	pH	–	2.85	7.42	5.50	1.93	35.09
		EC	mS/cm	12.40	24.11	17.59	3.72	21.13
		Sal	ppt	7.10	14.67	10.38	2.40	23.10
		LOI	%	2.34	14.07	6.54	3.50	53.54
		Sand	%	4.65	56.58	19.26	16.93	87.89
		Silt	%	37.42	74.41	58.93	10.64	18.05
		Clay	%	4.40	42.00	21.81	11.39	52.21
		Hg	μg/kg	16.55	61.12	39.33	12.91	32.83
	Water	pH	–	7.26	7.85	7.64	0.22	2.92
		EC	mS/cm	19.21	40.53	35.43	7.62	21.51
		Sal	ppt	11.30	25.83	22.29	5.22	23.41
Southern	Sediment	pH	–	3.45	7.75	6.38	1.52	23.76
		EC	mS/cm	8.92	33.50	16.32	4.96	30.39
		Sal	ppt	4.97	21.03	9.60	3.21	33.45
		LOI	%	3.47	14.76	8.02	3.27	40.81
		Sand	%	1.05	70.04	15.85	19.97	126.00
		Silt	%	21.99	80.46	52.67	16.48	31.28
		Clay	%	6.40	62.00	31.41	16.62	52.91
		Hg	μg/kg	32.54	89.05	64.59	16.09	24.91
	Water	pH	–	6.32	8.44	7.66	0.73	9.55
		EC	mS/cm	19.67	49.03	40.20	8.36	20.79
		Sal	ppt	11.67	32.20	25.76	5.80	22.52

SD standard deviation, *CV* coefficient of variation

of seawater tend to have higher pH, EC, and salinity values.

The pH values for the water samples ranged from 6.32±0.01 (almost neutral) at station 43 to 8.44±0.01 (alkaline) at station 31, while the pH values for the sediment samples ranged from acidic at station 27 (2.85±0.01) to alkaline at station 5 (7.97±0.01). For each sampling station, the EC for the water and sediment were significantly different from each other ($p<0.05$; ANOVA). However, among all the selected physico-chemical parameters under study, the EC (sediment) had a strong positive correlation ($r=0.999$; $p<0.05$) with salinity (sediment) (Table 4). The lowest EC (15.43±0.17 mS/cm) and salinity (7.43±0.06 ppt) for water were found at station 12 while the highest EC (49.03±0.06 mS/cm) and salinity (32.2±0.00 ppt) were found at station 33. As for sediment, the highest EC (46.37±0.06 mS/cm) and salinity (30.13±0.06 ppt)

Table 6 Concentration of Hg in sediments of Strait of Malacca relative to other regions

Region	Hg (μg/g)	Digestion method	Reference
Sundarban mangrove wetland, India	0.032–0.196	750 °C pyrolysis	Chatterjee et al. (2012)
Mangrove wetlands, China	0.026–0.468	HNO_3, H_2SO_4, 5 % $KMnO_4$	Ding et al. (2009)
Off Kuala Terengganu, Malaysia	0.020–0.127	HNO_3, H_2SO_4, HClO	Kannan and Falandysz (1998)
Anadyr Estuary, Russia	0.077–2.100	HNO_3, H_2SO_4, HClO	Kannan and Falandysz (1998)
Minamata Bay, Japan	1.400–4.300	HNO_3/HCl (1:1) and conc. H_2SO_4	Tomiyasu et al. (2006)
Fukuro Bay, Japan	0.300–4.800	HNO_3/HCl (1:1) and conc. H_2SO_4	Tomiyasu et al. (2006)
Dora, Lebanese coast, Estern Mediterranean	0.100–0.650	550 °C decomposition	Abi-Ghanem et al. (2011)
Amba estuary, India	0.050–2.660	Aqua regia and oxidation by $KMnO_4$	Ram et al. (2009)
Kaohsiung River mouth, Taiwan	0.150–1.150	HNO_3/HCl/HF (5:2:5)	Chen et al. (2012)
Salt marshes Bay of Fundy, eastern Canada	0.007–0.079	HNO_3/H_2SO_4 (7:3)	Hung and Chmura (2006)
Northern Gulf of Mexico	0.010–0.200	Modified EPA method 245	Apeti et al. (2012)
Continental coast of Shanghai	0.000–0.466	Aqua-regia	Deng et al. (2013)
Jade Bay, Southern North Sea	0.008–0.243	Oygen combustion-gold amalgamation	Jin et al. (2012)
Hongfeng Reservoirs, Southwest China	0.080–1.030	Microwave digestion (HNO_3-HF-$HClO_4$-HCl)	Wu et al. (2014)
Baihua Reservoirs, Southwest China	ND-2.200	Microwave digestion (HNO_3-HF-$HClO_4$-HCl)	Wu et al. (2014)
Strait of Malacca, Malaysia	0.017–0.114	Ultrasound extraction (HNO_3, KCl)	Current study

ND not detected

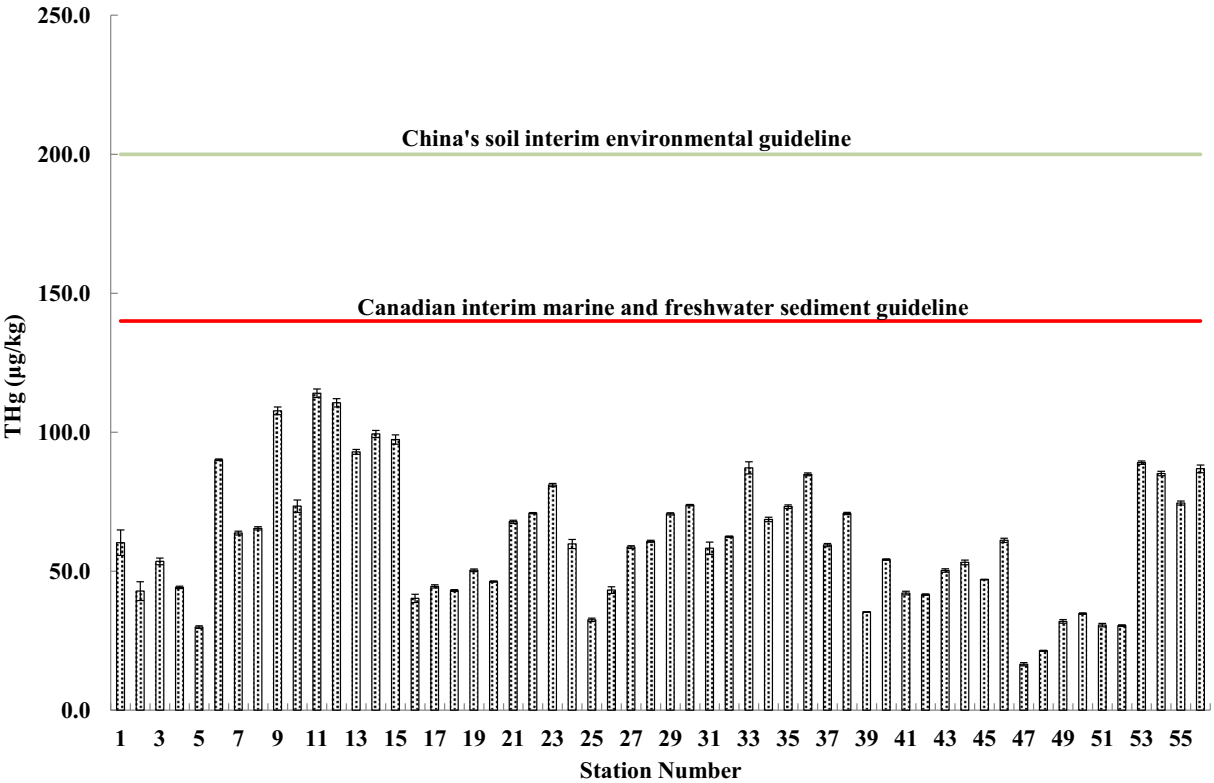

Fig. 4 Total mercury concentrations for estuaries and coastal sediments of the Strait of Malacca as compared to guidelines

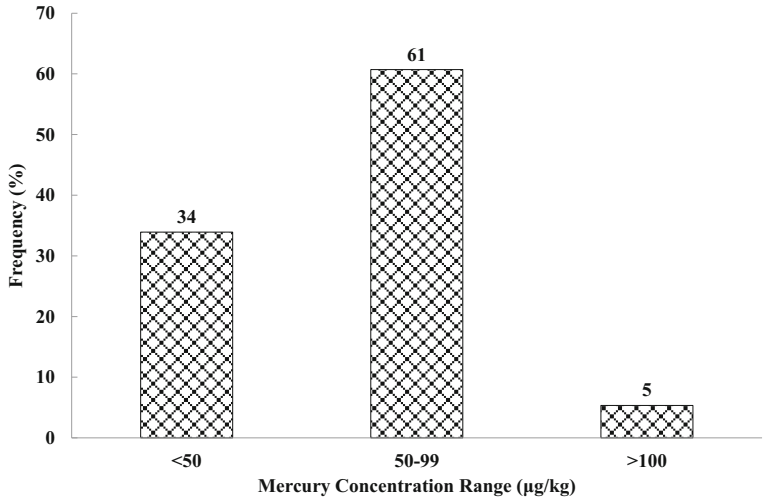

Fig. 5 Frequencies of mercury concentrations in estuary and coastal sediments of Strait of Malacca

were found at station 9 while the lowest EC (8.92± 0.30 mS/cm) and salinity (4.97±0.21 ppt) were found at station 52. In water, dissolved salts will dissociate, as the positively and negatively charged ions are capable of conducting an electric current (Mustapha et al. 2013). Thus, the significant relationship between EC and salinity was attributed to the presence of dissolved salts in the estuarine environment. Meanwhile, statistically significant differences were found between physico-chemical parameters of water and sediment to mercury (Hg) concentration (p values <0.05). From the result of correlation analysis, significant moderate negative correlations were found between Hg versus EC of water (r=−0.361; p values <0.01) and Hg versus salinity of water (r=−0.378; p values <0.01). However, there were significantly no correlation found between Hg versus EC of sediment (r=0.179; p values <0.05) and Hg versus salinity of sediment (r=0.187; p values <0.05). Nevertheless, no significant correlations were found between Hg versus pH of water samples (r=0.097; p values >0.05) and Hg versus pH of sediment samples (r=−0.079; p values >0.05) (Table 4).

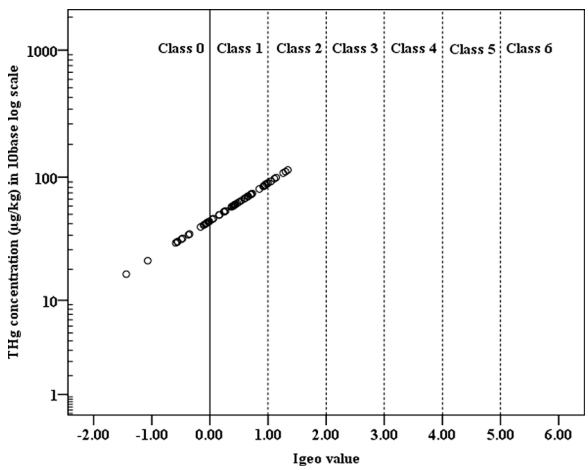

Fig. 6 Distribution of total mercury (THg) concentrations and I_{geo} values in estuaries and coastal sediment samples of the Strait of Malacca

Apart from that, LOI analysis revealed that the contents of organic matter vary (CV>30 %) along the Strait of Malacca. The highest percentage (14.76 %) of organic matter was recorded at station 53 whereas the lowest (2.34 %) at station 44. Basically, the metal concentration of sediment was influenced by the amount of organic matter present in the samples (Raj et al. 2013). In this study, the significant moderate (r=0.510; p<0.01) relationship of organic matter toward Hg concentration can be proved by Pearson correlation analysis. In addition, organic matter content in sediment samples were found to be influence by the supply rate of in-land materials, the rate of decomposition, and the texture and grain size of sediment (Raj et al. 2013). The texture control over organic matter is indicated by positive moderate correlation of LOI to fine-grain sediment, especially silt (r= 0.317; p<0.01) (Table 4). Similarity in settling velocity causes co-sedimentation of organic matter with fine grains. As such, organic coating onto fine grain may greatly affect the distribution of inorganic elements in the sediment samples (Raj et al. 2013). Particle size analysis demonstrated that more than 90 % of total sampling stations are predominated by fine-grain sediment. Out of 56 sampling stations, 51 sampling locations had fine-grain (<50 μm) content higher than 50 % (Fig. 3). The highest percentage (98.95 %) of fine-grain

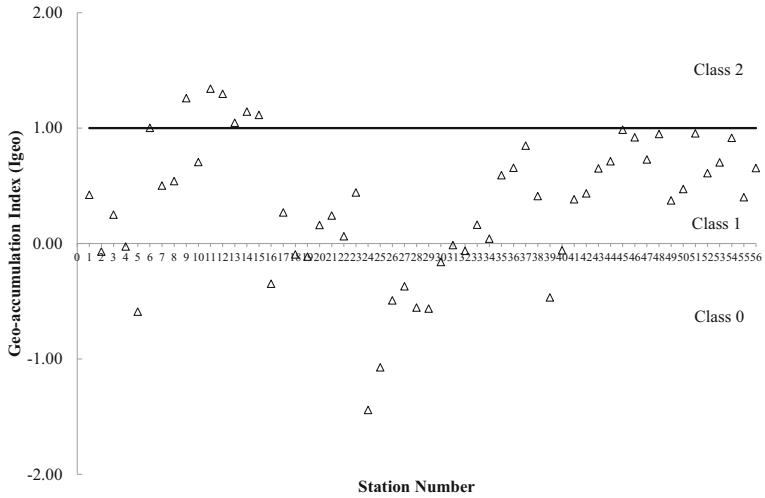

Fig. 7 Geoaccumulation Index (I_{geo}) for sediments collected from 56 sampling stations along the Strait of Malacca

sediment was recorded at station 45 while the lowest (29.99 %) was detected at station 55. Basically, sediment particle sizes are varies along the coast of the Strait of Malacca. Factor such as land-based run-off is contributing to the transportation of suspended solid and governed sedimentary process in the study areas (Sany et al. 2012).

Mercury concentration in estuaries and coastal sediment

The high variability of Hg concentrations in the estuaries and coastal sediments of the Strait of Malacca can be observed from the high CV value (37.85 %). The highest mercury concentration (114.02±1.54 μg/kg) was recorded in the sediment collected from Kuala Juru (station 11) whereas the lowest mercury concentration (16.55±0.61 μg/kg) was detected at station 24. Indeed, the Juru River was the most polluted river in Malaysia (Lim and Kiu 1995; Al-Shami et al. 2010) due to the pollution input from various urban and industrial activities (Yap and Tan 2008). Al-Shami et al. (2010) pointed out that the Department of Environment (DOE) has categorized the Juru River as "very polluted" according to its water quality index (WQI). However, the Juru River water quality has improved from class IV (2004) to class III (2006) within 2 years after considerable effort and good management practices were implemented to clean up and rehabilitate the river (Department of Environment DOE 2007). Therefore, the present Hg concentration was low compared to the mercury concentration (201 μg/kg) reported by Yap et al. (2003b) in the year 1999.

The comparative study of the current Hg concentration in sediments between the three parts—northern, central, and southern—of the west coast of Peninsular Malaysia showed that the northern part had the highest mean Hg concentration (76.36±27.25 μg/kg), followed by the southern (64.59±16.09 μg/kg) and central (39.33 ±12.91 μg/kg) parts (Table 5).

This finding is in accordance with the preliminary research done by Yap et al. (2003b) in 1999. The comparison of Hg concentration found that there are statistically significant differences between the northern and southern parts compared to the central part ($p<0.05$; Tukey-Kramer test). However, there was no statistically significant difference in the Hg concentration between the northern and southern parts ($p>0.05$; Tukey-Kramer test). Apart from station 11, high levels of Hg were also recorded in the sediment samples collected from station 12 (110.61± 1.51 μg/kg) in the Juru River estuary and station 9 (107.71±1.35 μg/kg) in Tanjung Dawai, north Peninsular Malaysia. The high levels of Hg found in the northern and southern parts were about one to two times higher than those found in the central part of Peninsular Malaysia. The elevated Hg mercury concentration in station 11 (highest in the northern part) was about seven times higher than that found in station 24 (lowest in the central part). This is probably due to station 11 being close to the Prai Industrial Estate (Yap et al. 2003b) while station 24 was protected from mercury accumulation due to the high velocity of water in this area (Haris and Aris 2012).

In a comparison work, the concentration of Hg detected in current studies was found to be relatively lower compared to sediment samples collected from other regions such as China, India, Japan, eastern

Mediterranean, and Taiwan (Table 6). Indisputably, estuary sediments are feasible places for the sink of Hg, still their abundance are influenced by factors like the distance to the proximity city, economic level, industrialization, land-based contaminants, and geomorphic and hydrological properties. For instance, mangrove sediments samples collected near big cities of Sanya, Shenzhen, Fugong, and Quanzhou of China have higher mercury pollution level (Ding et al. 2009). In addition, historical pollution event might even influence the abundance of Hg in the environment. It has already been about half a century since the outbreaks of the Minamata disease. Today, the Hg concentration found in Minamata Bay is still high compared to other regions of the world. However, the Hg concentrations in the sediment samples under study were all below the Canadian interim marine and freshwater sediment guideline (140 μg/kg) (Gaudet et al. 1995) and China's soil interim environmental guideline (200 μg/kg) (Wu et al. 1991) (Fig. 4).

In spite of the fact that Hg concentrations in the Strait of Malacca were found to be in lower orders of magnitude than the Hg concentration levels in others regions, it is undeniable that the Hg enrichment could occur in any circumstances.

According to Sadiq (1992), 50.0 μg/kg of Hg concentration can be used as background mercury concentration in uncontaminated sediment. Generally, 66 % of estuaries and coastal sediment collected from the Strait of Malacca had a Hg concentration that exceeded the background mercury level (Fig. 5.). Only 34 % of all the sampling stations had <50 μg/kg of Hg in their sediment samples.

In order to have a better and true reflection of the mercury enrichment in the sediment of the Strait of Malacca, I_{geo} was used as a reliable tool to compare the current Hg concentration to its background value from a previous study (Haris and Aris 2012). In general, Hg concentrations in sediment samples had a strong positive correlation between its I_{geo} values (r=0.969; p<0.05). This explains that the higher I_{geo} values were driven by the higher concentrations of Hg found in the sediment samples (Fig. 6).

From Fig. 7, about 13 % of all the sampling stations (station 6, 9, 11–15) along the Strait of Malacca had moderately contaminated Hg (class 2). These stations covered the sampling stations from Tanjung Dawai, Kedah, to Kuala Juru, Penang, which are located in the northern part of Peninsular Malaysia. According to Hajep and Jinap (2011), communities from Tanjung Dawai had the highest seafood consumption (655.7 g/person/kg) among the four coastal states (Johor, Kedah, Terrengganu, and Selangor) in Peninsular Malaysia. The mercury intake for this population was reported as being 2.254 μg/kg body weight/day (Hajep and Jinap 2011). Therefore, in relation to the enrichment of Hg in Tanjung Dawai, the population from this area, especially the fisherman communities who have the highest fish ingestion and mercury exposure, are at risk. However, about 30 % of the total number of sampling stations were practically uncontaminated by mercury and classified as class 0 in I_{geo} classification and the remaining 57 % of sampling stations had uncontaminated to moderately contaminated (class 1) Hg concentration.

Conclusion

The flow injection mercury system used in this study for Hg determination is as sensitive as the previous study. The findings from this study showed that the northern and southern parts of Peninsular Malaysia had a higher Hg concentration in the sediments compared to those found in the central part. However, the Hg concentrations detected in the present study were all below 140 μg/kg (Canadian interim marine and freshwater sediment guideline). I_{geo} successfully revealed that a few sampling stations from Tanjung Dawai and Kuala Juru were experiencing moderate Hg enrichment while most of the sampling sites along the Strait of Malacca were classified as uncontaminated to moderately contaminated sites. By taking into account that the bioavailable phase of mercury in sediment could contribute to mercury toxicity in aquatic organisms, periodically monitoring Hg concentrations in an aquatic environment is essential with special focus on the coast near to the industrial areas and its proximity. Therefore, the current Hg profiling will provide good insights for future mercury pollution and monitoring studies along the Strait of Malacca. In addition, it will benefit the relevant authorities in reviewing policies and managing environmental mercury pollution.

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