Bull Environ Contam Toxicol (2010) 84:568–573
DOI 10.1007/s00128-010-9998-8

Johor Strait as a Hotspot for Trace Elements Contamination in Peninsular Malaysia

Syaizwan Zahmir Zulkifli · Ahmad Ismail · Ferdaus Mohamat-Yusuff · Takaomi Arai · Nobuyuki Miyazaki

Received: 14 October 2009 / Accepted: 8 April 2010 / Published online: 22 April 2010

Abstract Present study was conducted to evaluate current status of trace elements contamination in the surface sediments of the Johor Strait. Iron (2.54 ± 1.24%) was found as the highest occurring element, followed by those of zinc (210.45 ± 115.4 µg/g), copper (57.84 ± 45.54 µg/g), chromium (55.50 ± 31.24 µg/g), lead (52.52 ± 28.41 µg/g), vanadium (47.76 ± 25.76 µg/g), arsenic (27.30 ± 17.11 µg/g), nickel (18.31 ± 11.77 µg/g), cobalt (5.13 ± 3.12 µg/g), uranium (4.72 ± 2.52 µg/g), and cadmium (0.30 ± 0.30 µg/g), respectively. Bioavailability of cobalt, nickel, copper, zinc, arsenic and cadmium were higher than 50% of total concentration. Vanadium, copper, zinc, arsenic and cadmium were found significantly different between the eastern and western part of the strait ($p < 0.05$). Combining with other factors, Johor Strait is suitable as a hotspot for trace elements contamination related studies.

Keywords Johor Strait · Sediments · Trace elements · Bioavailability · Hotspot

S. Z. Zulkifli (✉) · A. Ismail (✉)
Department of Biology, Faculty of Science, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
e-mail: syaizwan@science.upm.edu.my

A. Ismail
e-mail: aismail@science.upm.edu.my

S. Z. Zulkifli · N. Miyazaki
Center for International Cooperation, Ocean Research Institute, The University of Tokyo, 1-15-1 Minamidai, Nakano-ku, Tokyo 164-8639, Japan

F. Mohamat-Yusuff
Department of Environmental Sciences, Faculty of Environmental Studies, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

F. Mohamat-Yusuff · T. Arai
International Coastal Research Center, Ocean Research Institute, The University of Tokyo, 2-106-1, Akahama, Otsuchi, Kamiheigun, Iwate 028-1102, Japan

High contamination of some trace elements in the environment have caused environmental concern related to their toxicity, prolonged persistence, bioaccumulation in organisms, and ultimately their effect on the aquatic environment and human health (Amin et al. 2009; Cuong and Obbard 2006; Long et al. 1995). Elevation of the levels of these pollutants in the vicinity of the Malacca Straits has been reported to originate from land-, sea- and atmospheric sources (Zulkifli et al. 2010; Ismail et al. 1993; Ismail and Rosniza 1997). These pollutants are eventually deposited into the sediment and biota of the aquatic environment.

Sediments are the ultimate sink for the numerous anthropogenic chemical contaminants that may be contained in effluents originating from agricultural, industrial, urban and recreational activities (Apitz et al. 2005). Bryan and Langston (1992) have recommended the use of sediment as a tool to assess impact of human activities on the aquatic environment. Locally in Peninsular Malaysia, Johor Strait is an important area because of the existence of mangroves, sea grasses, corals and mudflats ecosystems. It is also important in other aspects. These include the high population [estimated at around 3 million people (DSM 2002) and expected to increase sharply as soon as this area develops rapidly], – fish- and shellfish resources, being a major aquaculture producer with output in excess of 150,000 tonnes in 2005, presence of major ports (Tanjung Pelepas: ranked 18th in 2007 global TEU; Johor Port: ranked 82nd in 2007 global TEU; Tanjung Langsat: a new petrochemical port), and the latest of the new and larger economic zone (Iskandar Malaysia). These features are expected to enhance and accelerate "high-impact" development (industrial, urban, agriculture, etc.) in this area. As for the city-state of Singapore, much of the lands have developed into industrial-, urban-, agriculture- and aquaculture areas. Massive development along both sides of the strait could lead to significant physical- and chemical changes in the environment. There are several studies including those of DOE (2007), Goh and Chou (1997), Ismail et al. (1995), Ismail (2008), Sin et al. (1991), Shazili et al. (2006), and Wood et al. (1997) that report elevated levels of heavy metals on both sides of straits due to past human activities. Due to its strategic location, ecological importance, promise of development in the future and past history of heavy metal contamination, this study was conducted to evaluate the suitability of the Johor Straits is a potential hotspot for trace elements contamination related studies.

Materials and Methods

Samplings for sediment samples were conducted in 2008 and involved five sampling locations along the Johor Strait (Fig. 1). Sediment samples of the top 3–5 cm of the surface were collected in triplicate at the respective sampling sites. Each sediment sample was placed in an acid-washed polyethylene bag and kept frozen (−20°C) prior to further analytical analysis. Analytical procedures used in present study were as the following. The reagents used were all of analytical-reagent grade certified for high-purity. Samples were dried in the oven at 60°C for at least 1 day until a constant dry weight was obtained. Afterwards, the samples

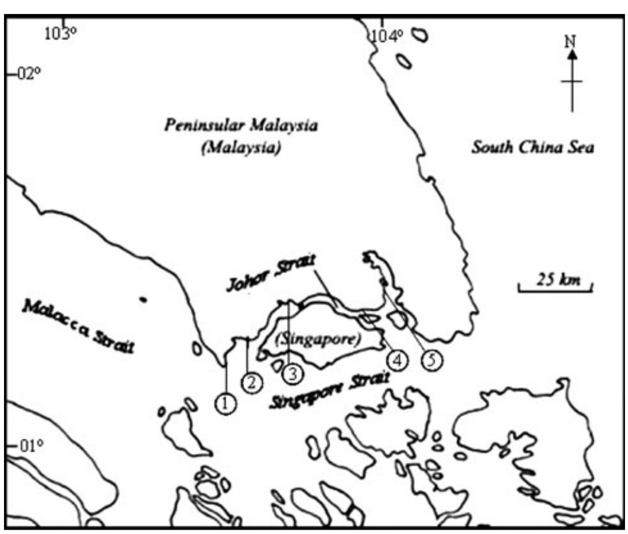

Fig. 1 Sampling locations along the Johor Strait

were agitated with a ceramic mortar and pestle. The fine fraction was obtained after sieving through a 63-µm mesh sieve. All sieved samples were kept in sealed acid-washed polyethylene containers prior to chemical analysis. To determine pseudototal trace elements content in sediments, the aqua regia digestion method was used according to ISO Standard 11466 (ISO 1995; Zulkifli et al. 2010). Briefly, 1.0 g of fine fraction sample was weighed and placed into the 250 mL Erlenmeyer flask. Next, 0.5–1.0 mL Mili-Q water (water resistivity >18.2 MΩ cm at 25°C; water TOC less than 5) was added to form a slurry. Then, 7.0 mL of 12 M HCl followed by 2.3 mL of 15.8 M HNO_3 (3:1 ratio) were added drop-by-drop to reduce foaming. The sample was allowed to pre-digest for 16 h (overnight). Afterwards, the suspension was digested on a hot plate at 130°C for 2 h, in an open system. The obtained suspension was then filtered through a 0.45 µm GD/X™ syringe filter into a polyethylene bottle. Finally, the filtered suspension was diluted to 50 mL with 0.5 M HNO_3 and stored at 4°C prior to metal content determination. In addition, the BCR (Community Bureau of Reference) sequential extraction procedure (Quevauviller et al. 1997) was conducted to fractionate metals or other elements in solid materials into several groups of differing leachability.

All samples treated with aqua regia and BCR sequential extraction methods were analysed using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500cs) for determining elemental content (^{51}V, ^{52}Cr, ^{59}Co, ^{60}Ni, ^{65}Cu, ^{66}Zn, ^{75}As, ^{107}Ag, ^{111}Cd, ^{208}Pb and ^{238}U). For qualitative measurements, Scandium (Sc), Indium (In) and Bismuth (Bi) were used as the internal standards, and the ICP Multi Element Standard Solution VI CertiPUR® was used as the external standard. All data are expressed on a dry weight basis (µg g^{-1} dry weight). Recoveries of trace elements ranged from 93.21% to 108.40% (ISO 11466) and 90.32% to 103.45% (BCR sequential extraction), respectively, using the Standard Reference Material® 1646a Estuarine Sediment (National Institute of Standards & Technology, USA). Enrichment factor (EF) was used to evaluate the actual extent of sediment contamination (Amin et al. 2009; Groengroeft et al. 1998), identify the element source between anthropogenic and naturally occurring (Morillo et al. 2004), and determine the degree of sedimentation (Huang and Lin 2003) using the formula:

$$\mathrm{EF} = \frac{(C_x/C_{\mathrm{Fe}})_{\mathrm{sample}}}{(C_x/C_{\mathrm{Fe}})_{\mathrm{shale}}}$$

where, C is concentration of element, x is type of element, $(C_x / C_{\mathrm{Fe}})_{\mathrm{sample}}$ and $(C_x / C_{\mathrm{Fe}})_{\mathrm{shale}}$ are element to Iron ratios in the sample and average shale level, respectively. Element concentration (µg/g dry weight) is expressed in relation to Fe levels (% dry weight) in sediment samples and the average shale values in Johor Strait were taken from Wedepohl (1971). Fe was used as it has a reliable association with fine solid surface, its geochemistry characters are similar to many trace elements, and its natural sediment concentration tends to be uniform (Daskalakis and O'Connor 1995).

Results and Discussion

In the present study, 11 elements were analyzed (V, Cr, Fe, Co, Ni, Cu, Zn, As, Cd, Pb, and U). Among these elements, the highest mean concentration was of Fe (2.54 ± 1.24% dry weight), followed by those of Zn (210.45 ± 115.4 µg/g), Cu (57.84 ± 45.54 µg/g), Cr (55.50 ± 31.24 µg/g), Pb (52.52 ± 28.41 µg/g), V (47.76 ± 25.76 µg/g), As (27.30 ± 17.11 µg/g), Ni (18.31 ± 11.77 µg/g), Co (5.13 ± 3.12 µg/g), U (4.72 ± 2.52 µg/g), and Cd (0.30 ± 0.30 µg/g), respectively. These results also reveal that the trace element concentrations in Johor Strait are within the ranges reported in other studies carried out around Peninsular Malaysia and other parts of the world (Table 1). All the studies have emphasised the input of anthropogenic trace elements into the environment. Comparisons of element concentrations between the eastern and western sides of the Johor Strait showed that the east has higher concentration of analyzed elements than the west, except for Co. There was also a significant difference in V, Cu, Zn, As, and Cd concentrations between the two zones ($p < 0.05$). These results correspond well with results of Cuong et al. (2005), which suggested that samples from the eastern side of the Johor Strait should be 1–5 times more contaminated than those of the western side, depending on the elements and specific sampling locations. Yap et al. (2006) also reported the sediment from Kg. Pasir Puteh (JPP) was more polluted with Cd, Cu, Ni, Pb, and Zn compared to sediment from Tanjung Piai (JTPi). Analyses using biological samples obtained from different parts of this strait also reveal a similar pattern. Yap et al. (2006) considered eastern parts of the Johor Causeway more polluted than the western after a sample of *Perna viridis* was analysed and was followed by analyses of samples from Senibong, Telok Jawa, Kg. Masai and Kg. Pasir Puteh which contained higher Cd, Cu, Fe, Pb, Ni, and Zn compared to samples collected from Kg Sg Melayu, Kg Gelang Patah dan Pantai Lido. Bayen et al. (2004) also reported the east of Johor Strait was found to be more contaminated with trace elements than the west when they used *P. viridis* as a bioindicator. Wood et al. (1997) reported that Johor Strait is contaminated by trace elements such as Cu, Zn, Pb, and Cd. The presence of anoxic sediments along both sides of the Johor Causeway generally confirms the severe contamination occurring in the area. High levels of Pb and Zn found near the causeway were attributed to anthropogenic inputs such as vehicles traffic across the

Table 1 Mean concentration of trace elements (µg/g dry weight) in locations along the Johor Strait and other areas around Peninsular Malaysia

Location	V	Cr	Fe[a]	Co	Ni	Cu	Zn	As	Cd	Pb	U	Reference
Western Johor Strait	13.3–66.1	12.6–81.9	0.8–3.6	1.6–10.0	2.8–24.7	2.0–37.5	29.4–225.3	0.2–41.4	0.02–0.2	8.7–66.1	0.7–7.3	Present study
Eastern Johor Strait	60.7–75.1	28.3–84.8	1.6–3.8	2.6–6.8	8.4–34.2	88.9–147.0	116.8–606.0	29.2–45.8	0.3–1.0	44.7–91.8	3.6–7.0	Present study
Pulau Tekong and Kranji	–	40.6–47.3	–	–	17.1–26.1	7.7–17.9	49.8–62.1	–	0.06–0.19	26.1–29.8	–	Cuong and Obbard (2006)
Sungei Buloh and Sungei Khatib Bongsu	–	16.61–32.07	–	–	7.44–11.65	7.06–32.00	51.24–120.23	–	0.18–0.27	12.28–30.98	–	Cuong et al. (2005)
Straits of Johor (average)	72.5	45.2	3.04	5.8	30.2	30.7	132.5	19.7	0.18	42.3	5.2	Wood et al. (1997)
Malaysian coast	–	–	–	–	–	1.79–8.17	13.86–42.43	–	0.27–0.54	6.16–27.52	–	Ismail et al. (1995)
Juru	–	–	1.90	–	–	29	84	–	–	25	–	Lim and Kiu (1995)
West coast of Peninsular Malaysia	–	–	2.50–5.00	–	–	$<$6.00	–	–	–	1.00–45.00	–	Ismail et al. (1993)

Remark: *ND* not detected; [a] % dry weight causeway and industrial discharges nearer shore. Furthermore, any disturbance by sea currents or human activities will cause resuspension of the sediment-bind-trace elements in the environment and probably in the soluble forms readily absorbed by aquatic organisms. A study by Nayar et al. (2004) indicated intensive dredging, reclamation, construction, and shipping activities in the Ponggol Estuary of Singapore (east of Johor Strait) may have led to resuspension and bioavailability of particulate elements. They reported presence of Sn, Pb, Ni, Cd and Cu in particulate- and dissolved fractions and sediments whose were ranges were from n.d.-92 μg/g, n.d.-303.2 μg/g, n.d.-2,818.4 μg/g, n.d.-74.4 μg/g and n.d.-1,117.7 μg/g, respectively. Consequently, toxic elements released from sediments may prove poisonous to the organisms that may have accumulated including bacteria (Nayar et al. 2004).

The present study also confirmed element concentrations derived from both anthropogenic- and lithogenic sources. The three-step BCR sequential extraction of the European Community Bureau of Reference was adopted for determining the sources and distribution of elements in different phases, including of the anthropogenic sources (through the exchangeable/carbonate fraction, easily reducible fraction and oxidisable fraction) and of lithogenic sources. The results obtained are shown in Fig. 2. Generally, more than 50% of Co, Ni, Cu, Zn, As, and Cd in the surface sediment samples originated from anthropogenic sources. Element concentration patterns in east and west Johor Strait were slightly different from each other, particularly for Cr and U. The anthropogenesis of these elements should be recent. They are said to enter via riverine inputs, non-point source runoffs from land and direct point source discharges. Most rivers make a major contribution to trace element loading in estuaries and coastal waters. Significant amounts of some trace elements found in river waters were reported to be from weathering of rocks and leaching of soils. Most notable anthropogenic sources of trace elements are from developed zones along rivers and coastal areas. Yap et al. (2006) reported higher anthropogenic input of trace elements in surface sediment samples collected from the Pasir Gudang area compared to other areas in the Straits of Malacca. As for the marine water quality analysed by DOE (2007), major contributors to the changes in its quality are from total suspended solids followed by oil and grease, e-coli, and metals such as Cu, Pb, Hg, and Cd. High levels of activity in industry in Johor

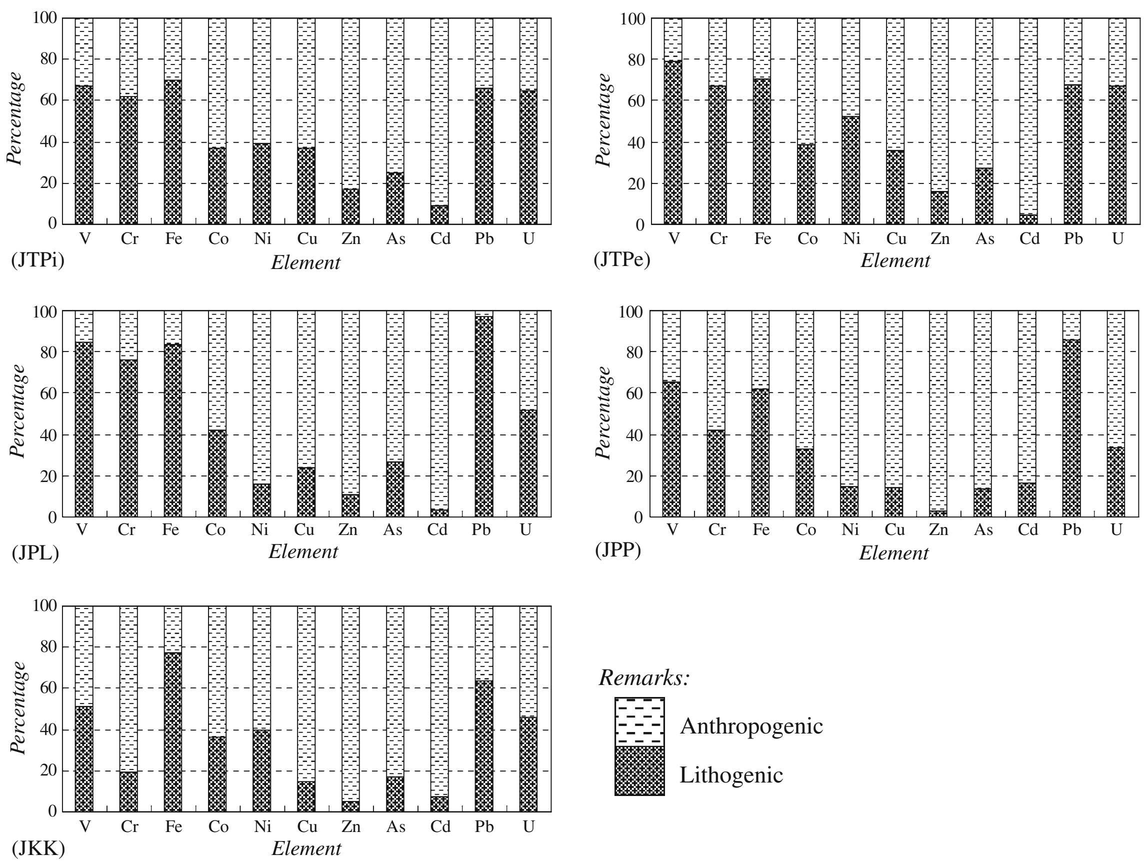

Fig. 2 Anthropogenic and lithogenic sources of trace elements in sediments collected along the Johor Strait

Table 2 Enrichment factors of trace elements in the Johor Strait

Element	V	Cr	Co	Ni	Cu	Zn	As	Cd	Pb	U
Area										
JTPi	1.30[a]	2.33[a]	1.34[a]	0.81	0.55	2.02[a]	10.61[c]	0.32	6.56[b]	5.03[b]
JTPe	0.26	0.36	0.21	0.11	0.38	0.79	0.04	0.06	1.11[a]	0.48
JPL	0.46	1.99[a]	0.64	0.93	1.86[a]	6.07[b]	5.11[b]	0.66	8.46	3.48[d]
JPP	1.20[a]	2.41[a]	0.91	1.29[a]	5.57[b]	16.33[c]	11.72[c]	3.03[d]	11.75[c]	4.85[d]
JKK	1.48[a]	0.80	0.35	0.31	8.18[b]	3.15[d]	7.47[b]	0.83	5.72[b]	2.50[a]

Remarks: [a] minor enrichment; [b] moderately severe enrichment; [c] severe enrichment; [d] moderate enrichment

contribute to air pollution including suspended particular matter (SPM). SPM may deposit in the coastal areas carrying pollutants as trace elements. Atmospheric depositions also may have contributed, even though in less significant amounts, to trace elements in the aquatic environment. Zhang et al. (2007) reported atmospheric transport of trace elements has been found to be an important pathway for their input into the ocean including coastal marine environment. In all of Malaysia, Johor was among the high contributors of air pollution sources after Selangor in 2006 (DOE 2007). Wu and Ting (2006) reported that fly ash from a municipal solid waste incinerator in Singapore was very fine (0.04–100 μm) in nature and contained a significant amount of toxic trace elements such as Al, Pb, Zn and others. Most of these major constituents are volatile elements which were formed due to high temperature and excellent burnout in the incineration process.

Table 2 shows the mean EF values of trace elements in the Johor Strait with respect to average shale (Wedepohl 1971). By analysing EF, the actual extent of sediment contamination can be assessed. The results showed that element enrichment factors in the sediment along the Johor Strait varied between no enrichment to moderately severe enrichment, depending upon sampling area and element. Generally, EF values showed agreement with both applied analytical analyses. Notably, locations in the eastern Johor Strait (JPP, JKK) have higher enrichment activities than the western part, particularly for Cu, Zn, Cd, and Pb. JKK could be suggested as the hotspot since severe enrichment of Cu, moderately high enrichment of Cd, and moderate enrichment of Zn and Pb were found. JPP was found to have only moderately severe enrichment of Cu and Zn, and moderate enrichment of Cd. Locations in western Johor Strait can be generally classified between no enrichment to minor enrichment. These results agree with past reports that the eastern side of the straits have more heavy metal pollution compared to the west (Bayen et al. 2004; Ismail 2008).

In general, most data from this study fall within the moderate and high levels of the range of metals reported in the region. Furthermore, the results showed high anthropogenic input of metals into the straits sediments. Continuous development may contribute to the progressive elevation of these elements. Therefore, we suggest that this area be declared a hotspot for trace elements contamination related studies in the Southeast Asia region. Future intensive studies related to ecotoxicology, environmental impact, management, and conservation should be conducted frequently in this hotspot, in order to understand all physical-, chemical-, and biological processes connected with trace element accumulation in the coastal ecosystem.

Acknowledgments This study was supported by a grant-in-aid from Scientific Program (No. 19380224), the Multilateral Core University Program "Coastal Marine Science" from the Japan Society for Promotion of Science, and the eScienceFund (Project No. 06-01-04-SF0715) from the Ministry of Science, Technology and Innovation of Malaysia.

References

Amin B, Ismail A, Aziz A, Yap CK, Kamaruddin MS (2009) Anthropogenic impacts on heavy metal concentrations in the coastal sediments of Dumai, Indonesia. Environ Monit Assess 148:291–305

Apitz SE, Davis JW, Finkelstein K (2005) Assessing and managing contaminated sediments: part I. Developing an effective investigation and risk evaluation strategy. Integr Environ Assess Manag 1:2–8

Bayen S, Thomas GO, Lee HK, Obbard J (2004) Organochlorine pesticides and heavy metals in green mussel, *Perna viridis*, in Singapore. Water Air Soil Pollut 155:103–116

Bryan GW, Langston WJ (1992) Bioavailability, accumulation and effects of heavy metals in sediments with special reference to United Kingdom estuaries: a review. Environ Pollut 76:89–131

Cuong DT, Obbard N (2006) Metal speciation in coastal marine sediments from Singapore using a modified BCR sequential extraction procedure. Appl Geochem 21:1335–1346

Cuong DT, Bayen S, Wurl O, Subramanian K, Wong KKS, Sivasothi N, Obbard JP (2005) Heavy metal contamination in mangrove habitats of Singapore. Mar Pollut Bull 50:1713–1744

Daskalakis KDO, O'Connor TP (1995) Normalization and elemental sediment contamination in the coastal United States. Environ Sci Technol 29:470–477

DOE (2007) Annual Report 2006. Department of Environment, Kuala Lumpur

DSM (2002) Population distribution by local authority areas and mukims. Jabatan Perangkaan Malaysia, Kuala Lumpur

Goh BPL, Chou LM (1997) Heavy metal levels in marine sediments of Singapore. Environ Monit Assess 44:67–80
Groengroeft A, Jaehnig U, Miehlich G, Lueschow R, Maass V, Stachel B (1998) Distribution of metals in sediments of the Elbe Estuary in 1994. Water Sci Technol 37:109–116
Huang KM, Lin S (2003) Consequences and implication of heavy metal spatial variations in sediments of the Keelung River drainage basin, Taiwan. Chemosphere 53:1113–1121
Ismail A (2008) A need for monitoring of heavy metals and organotin compounds in the east coast of Johor. In: Che Abdul Rahim M, Masri MA, Zaidi CK, Norhayati A (eds) Research and information series of Malaysian coast of Malaysia marine ecosystem. Marine Ecosystem Research Centre UKM, Bangi, pp 163–176
Ismail A, Rosniza R (1997) Trace metals in sediments and molluscs from an estuary receiving pig farms effluent. Environ Technol 18:509–515
Ismail A, Badri MA, Ramlan MN (1993) The background levels of heavy metals concentrations in sediments from the west coast of Peninsular Malaysia. Sci Total Environ 134:315–323
Ismail A, Jusoh NR, Ghani IA (1995) Trace metal concentrations in marine prawns off the Malaysian coast. Mar Pollut Bull 31: 108–110
ISO (1995) ISO 11466: soil quality, extraction of trace elements soluble in aqua regia. International Organization for Standardization, Geneva
Lim PE, Kiu MY (1995) Determination and speciation of heavy metals in sediments of Juru River, Penang, Malaysia. Environ Monit Assess 35:85–95
Long ER, MacDonald DD, Smith SC, Calder FD (1995) Incidence of adverse biological effects within ranges of chemical concentrations in marine and estuarine sediments. Environ Manag 19: 81–97
Morillo J, Usero J, Gracia I (2004) Heavy metal distribution in marine sediments from the southwest coast of Spain. Chemosphere 55: 431–442
Nayar S, Goh B, Chou L (2004) Environmental impact of heavy metals from dredged and resuspended sediments on phytoplankton and bacteria assessed in in situ mesocosms. Ecotoxicol Environ Safe 59:349–369
Quevauviller P, Rauret G, López-Sánchez J-F, Rubio R, Ure A, Muntau H (1997) Certification of trace metal extractable contents in a sediment reference material (CRM 601) following a three-step sequential extraction procedure. Sci Total Environ 205:223–234
Shazili NAM, Yunus K, Ahmad AS, Abdullah N, Rashid MKA (2006) Heavy metal pollution status in the Malaysian aquatic environment. Aquat Ecosyst Health 9:137–145
Sin YM, Wong MK, Chou LM, Normala A (1991) A study of the heavy metal concentrations of the Singapore River. Environ Monit Assess 19:481–494
Wedepohl KH (1971) Environmental influences on the chemical composition of shales and clays. Phys Chem Earth 8:307–331
Wood AK, Ahmad Z, Shazili NAM, Yaakob R, Carpenter R (1997) Geochemistry of sediments in Johor Strait between Malaysia and Singapore. Cont Shelf Res 17:1207–1228
Wu HY, Ting YP (2006) Metal extraction from municipal incineration fly ash—chemical leaching and fungal bioleaching. Enzyme Microb Technol 38:839–847
Yap CK, Choh MS, Edward FB, Ismail A, Tan SG (2006) Comparison of heavy metal concentrations in surface sediment of Tajung Piai wetland with other sites receiving anthropogenic inputs along the southwestern coast of Peninsular Malaysia. Wetl Sci 4:48–57
Zhang X, Zhuang G, Guo J, Yin K, Zhang P (2007) Characterization of aerosol over the Northern South China Sea during two cruises in 2003. Atmos Environ 41:7821–7836
Zulkifli SZ, Mohamat-Yusuf F, Arai T, Ismail A, Miyazaki N (2010) An assessment of selected trace elements in intertidal surface sediments collected from the Peninsular Malaysia. Environ Monit Assess. doi:10.1007/s10661-009-1189-x