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Spatial assessment and source identification of heavy metals pollution in surface water using several chemometric techniques

Azimah Ismail [a], Mohd Ekhwan Toriman [a,*], Hafizan Juahir [a], Sharifuddin Md Zain [c], Nur Liyana Abdul Habir [a], Ananthy Retnam [b], Mohd Khairul Amri Kamaruddin [a], Roslan Umar [a], Azman Azid [a]

[a] East Coast Environmental Research Institute (ESERI), Universiti Sultan Zainal Abidin, Gong Badak Campus, 21300 Kuala Terengganu, Malaysia
[b] Environmental Health Division, Department of Chemistry Malaysia, Ministry of Science, Technology and Inovation, Jalan Sultan, Petaling Jaya, Selangor, Malaysia
[c] Chemistry Department, University of Malaya, 50603 Kuala Lumpur, Malaysia

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ABSTRACT

This study presents the determination of the spatial variation and source identification of heavy metal pollution in surface water along the Straits of Malacca using several chemometric techniques. Clustering and discrimination of heavy metal compounds in surface water into two groups (northern and southern regions) are observed according to level of concentrations via the application of chemometric techniques. Principal component analysis (PCA) demonstrates that Cu and Cr dominate the source apportionment in northern region with a total variance of 57.62% and is identified with mining and shipping activities. These are the major contamination contributors in the Straits. Land-based pollution originating from vehicular emission with a total variance of 59.43% is attributed to the high level of Pb concentration in the southern region. The results revealed that one state representing each cluster (northern and southern regions) is significant as the main location for investigating heavy metal concentration in the Straits of Malacca which would save monitoring cost and time.

Capsule: The monitoring of spatial variation and source of heavy metals pollution at the northern and southern regions of the Straits of Malacca, Malaysia, using chemometric analysis.

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1. Introduction

The increase in urbanization has led to an increase of pollutant discharge points into the Straits of Malacca, from both land-based and sea-based sources in a bulk-scale (Abdullah et al., 1999). Anthropogenic inputs contribute to the presence of pollutants that exhibit high toxicity into the marine ecological environment. The potential accumulation of waterborne heavy metals particularly Cadmium, Cuprum, Lead and Arsenic may have potential threat to the natural environmental marine communities and aggravate human health (Zakaria and Takada, 2007). In Malaysia, most pollutants are generated from industrial activities, intentional discharge from operation of vessels at ports, gas and crude oil platforms, petroleum refining and fertilizer manufacturing.

According to Department of Environment (DOE) (2008), extensive changes in land use and industrialization have cause proliferation of heavy metal wastes like cadmium (Cd), copper (Cu), lead (Pb) and mercury (Hg), specifically in the littoral states; Pulau Pinang, Perak, Selangor and Malacca into the Malacca Straits. Manufacturing sectors contribute a wide variety heavy metal pollutants in the coastlines of Western Peninsular Malaysia (Thia-Eng et al., 2000). Persistent organic pollutants (POPs) from the petroleum spills or tanker wreckage such as nickel and copper have environmentally contaminated and polluted the seawater surface and sediment, leading to the accumulation of heavy metals in aquatic systems (Santos-Echeandia et al., 2009). In addition, marine estuarine environment will be significantly impacted from accumulation of toxic substances and oil spill (Sun et al., 2012). Metal concentrations in sediment cores for example, were detected to be considerably higher in 1993, along the east–west axis of the Straits of Johore between Singapore and Malaysia compared to other years (Wood et al., 1997).

Heavy metals and metallic chemical elements (such as hydrogen, lithium,sodium, potassium and rubidium), highly dense and toxic, non-biodegradable substances are transported into marine ecological environment through storm runoff, and subsequently result in bioaccumulation in aquatic living organisms over a long period, dangerously affecting human health via consumption of food from a variety of aquatic life forms (Irwandi and Farida, 2009; Nazli and Hashim, 2010). Agricultural sector is another endocrine contributor of heavy metals in the environment, sourced from fertilizers, pesticides and herbicides. Research in marine environmental system relating to ecotoxicology stated that mercury, cadmium, copper and zinc are the most dangerous heavy metals in the environment (Golovanova, 2008). Studies on identification and apportionment of heavy metals in the environment have been conducted worldwide. One example is the study on the accumulation of mercury, chromium, lead, arsenic and cadmium in European

* Corresponding author.
E-mail address: ekhwan@unisza.edu.my (M.E. Toriman).

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catfish into human body via food webs, skin and all important organs on aquatic ecosystems, leading to potential deleterious effects to humans (Aquadrone et al., 2013). Cadmium and lead have been traced in living organisms such as littoral shore mollusk (Thais species), rock oyster and mangrove plant sediment in the West Coast of Peninsular Malaysia, along the Straits of Malacca (Shazili et al., 2006; Nazli and Hashim, 2010). It is unfortunate that economic development often leads to marine pollution by waste discharges from agricultural activities such as pesticide and fertilizers, industries such as heavy metals and hydrocarbon, sewage effluents and shipping such as an oil spillage, which consist of harmful substances that can affect human health and other organism, hence can change the quality of the seawater.

Chemometric techniques allow the study of statistical-correlation in terms of faunal structure, physicochemical characteristics and toxicological data, are found to be significant in analyzing dataset from laboratory analyses. The techniques provide the most appropriate tools for a meaningful data reduction and interpretation (Satheeshkumar and Khan, 2011). Moreover, the application of unbiased and precise methods viz. principal component analysis (PCA), cluster analysis (CA) and discriminant analysis (DA) and multi linear regression offers better insight and in-depth interpretation of heavy metals in water surface. Kannel et al. (2007) had suggested the application of a variety of methods for multivariate analyses to reduce the complexity of large amounts of data and provide better data interpretation. CA, as an unsupervised pattern recognition technique functions to group variables and observations. It is able to establish heavy metal signatures and evaluate potential sources of pollutant variables (Han et al., 2006). Further, CA helps in the identification of heavy metal fingerprint and its origin and variations between sampling sites along the Straits of Malacca. Commonly, the application of CA is to group the sampling sites of heavy metals having similar characteristics and to identify variations between sampling sites. Results obtained from the clustering enables data interpretation and pattern recognition of heavy metals. DA can be used to support CA as a pattern recognition (Juahir et al., 2011) and data classification technique with prior knowledge of the objects of the particular sampling sites (Singh et al., 2004). PCA is commonly applied as a data reduction technique that reduces the large amounts of variables into smaller set of components that can be easily interpreted (Zhou et al., 2007). The application of PCA provides important information that describes the whole data set without losing any of the original information. PCA is usually employed to detect correlation between heavy metal variables for source identification in water surface.

This study aims to identify heavy metal pollution distribution and the most significant sources via a comprehensive application of multivariate analyses, on data sets obtained from the Department of Environment (DOE). Datasets consist of a five-year period (2006–2010) marine water heavy metals concentrations. It is desired that results obtained from this study will enable the optimization of the monitoring program along the Straits of Malacca as a pollution control mechanism and source identification of heavy metal pollutants. Furthermore, the involvement of environmental forensics has the potential to assist relevant enforcement agencies to bring the polluters to justice and all related issues concerning pollution of heavy metals can be addressed more fruitfully.

2. Methodology

2.1. Study areas

This study was conducted along the Straits of Malacca, located at the West Coast of Peninsular Malaysia. The study area covers the coordinates ′N 06° 20′ to N 01° 41′ and E 100° 09′ to E 103° 05′, along the coastline areas of Perlis to Johor as shown in Fig. 1. The Straits of Malacca separates the Peninsular Malaysia and Sumatra of Indonesia. It forms a funnel-shaped waterway and narrows down towards the south. It ends in the area between Tanjung Piai, Johor, Malaysia and Pulau Karimun Kecil in Indonesia (Rusdi, 2012). The Malacca Straits has a tropical climate, which consists of wet and dry season and influenced by the northeast monsoon, bringing rain from December to February and dry season from June to August due to south-west monsoon. During the inter-monsoon period from March to May, and September to November, the weather is not stable and unpredictable (Thia-Eng et al., 2000).

The West Coast of Peninsular Malaysia is dominated by coastal plains and basin formed by alluvial deposits. The Malacca Straits is the busiest international shipping route in the world for crude oil trading from Middle East to Asian countries like China and South Korea (Zakaria and Takada, 2007). Since the last decade, states of Pulau Pinang and Johor are known to have robust urbanization activities. Both states are on a fast industrialization track, constantly upgrading their maritime shipping industries. Although the state of Johor is experiencing tremendous urbanization, the state remains as the most agricultural state in the West Coast of Malaysia, particularly palm oil and coconut plantations (Liow et al., 2001). Perak, which is also a state along the straits, has the largest tin extraction and mining activities in Malaysia.

2.2. Monitoring parameters and pre-treatment data

Datasets of marine water from eight monitoring station in the littoral states along the Straits of Malacca consist of nine parameters including six heavy metals obtained from the Department of Environment Malaysia (DOE) from 2006 until 2010. Sampling frequencies yearly are between two to nine times according to the areas. The industrialized and fully developed states such as Pulau Pinang require more than two sampling frequencies, while only two sampling frequencies were conducted in the least developed state like Perlis. For the analyses of heavy metals, we have focused on six parameters viz. arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb) and mercury (Hg). Prior to a comprehensive chemometric statistical analyses, an appropriate data checking viz. normal distribution and data transformation for each variable using a normality tests such as Shapiro–Wilk, Anderson–Darling, Liliforths and Jarque–Bera, with 95% confidence level is important. Chemometric techniques are sensitive to anomalies in data distribution especially when the raw dataset are not normally distributed ($p < 0.05$). Some of the data were below detectable limits and are non-numerical. Consequently, the dataset were transformed into a numerical form to ease the analyses (Junninen et al., 2004) using the following equations (Eqs. (1) and (2)):

$$Y = X – X/2 \text{ .... if it is noted} < X \tag{1}$$

$$Y = X/2 + X \text{ ....if it is noted} > X. \tag{2}$$

The data were transformed and arranged using log-transformation and z-scale standardized (mean = 0, standard deviation = 1) to ensure that each variables have the same influence in the analysis (Zhou et al., 2007; Satheeshkumar and Khan, 2011). After the data conversion, the dataset was formed into a single matrix of heavy metal variables vs. sampling points (littoral states), forming a [6 × 10] data matrix. The non-detectable data are replaced by one half of the detection limit value (Retnam et al., 2013).

The Z-scale transformation was applied using the following equation (Eq. (3)):

$$Z = \frac{x - x'}{\sigma} \tag{3}$$

where x is the original value of a measured parameter, Z is the standardized value, x' is the average value of variables and σ is the standard deviation (Kannel et al., 2007).
where, Y represents the estimated observation and X is the actual observation. The descriptive statistics, mean and standard deviation (S.D.) for

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