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Assessment of Heavy Metals in Sediment at Saphan Hin, Phuket

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This study was conducted to assess the level of metal pollution in sediment samples at two different locations at Saphan Hin, which received water from Bang-Yai canal. The total content of Tin (Sn), Lead (Pb), and Zinc (Zn) were measured using inductively coupled plasma optical emission spectrometer (ICP-OES) by aqua regia digestion. The ranges of metal concentration (mg kg^{-1}) were 9.04 - 32.17 mg kg^{-1} and 6.33 - 36.52 mg kg^{-1} of Pb in Core A and B; and 32.83 - 68.49 mg kg^{-1} and 21.63 - 73.59 mg kg^{-1} of Zn in Core A and B, which is below the Sediment Quality Guidelines of Threshold Effects Concentration (TEC) (35.8 mg Pb kg^{-1} and 121 mg Zn kg^{-1}). Sn was not detected (< 0.03 mg kg^{-1}). Analysis of the geo-accumulation index (I_{geo}) clearly indicated that most of the analysed sediment samples were not contaminated with the studied metals since low I_{geo} values were observed ($I_{geo\ Pb}$ and $I_{geo\ Zn}$ < 0). The detected enrichment factor (EF_{Pb}) values between 2.0 and 6.0 indicated moderate enrichment, while most of the sediments had EF_{Zn} values below 2 which exhibited deficiency to minimal enrichment. The EF values in this study (EF > 1.5) indicates a significant portion of the trace metals was delivered from non-crustal materials and that may be delivered by other sources. Correlation analysis showed positive relationships between Pb and Zn in both cores (r = 0.5** Core A and 0.941** Core B). The association of Pb and Zn is possibly because they originated from the same sources. The determination of other metals should be considered further.

1. Introduction

Heavy metals are major environmental pollutants and their toxicity is the cause of increasing concern due to their accumulation and persistence in the environment (Jaishankar et al., 2014). Metal distribution and transportation is considered to be significant and can also cause adverse effects to human health. The main sources of the heavy metals in the coastal environment are difficult to be clearly identified. High level of heavy metals can often be attributed to anthropogenic influences such as agricultural runoff, mining activities, industrial effluents, rather than natural enrichment by geological weathering (Goher et al., 2014). Sediments are the principal reservoir for heavy metals and act as indicator of the heavy metal burden in a coastal environment. The behaviour of metals in sediments reflects the historical deposition, sources of contaminants, and information on the impact of human activities on the coastal ecosystem (Sany et al., 2013).
Phuket is one of the southern provinces of Thailand and has been Thailand's most popular island as a tourism hub of the region. The discovery of Sn in Phuket began in 1909 as Tin mining in the Kathu district until 1992 when the last mine was closed (KoPhuket, 2016). Mineral resources are the most crucial elements for development throughout the world. Mining activities generated a large amount of waste rocks and tailings,

which results in heavy metal contamination in water sources, soils, and sediments that could contribute to the accumulation of heavy metals in environment. Bang-Yai canal is a primary waterway used to transport freight into Phuket town in the past. It has received water from the reservoir upstream at Kathu waterfall through ex-Tin mining area and Phuket Town into the sea coast at Saphan Hin. Bang-Yai canal can be considered as a potentially significant source of heavy metal pollution, especially with the urban expansion and economic development in the area and brought the risk of heavy metal contamination. The area is now becoming increasing polluted with organic and inorganic contaminant discharged into the canal. High concentration of Sn and Pb in the sediments from Bang-Yai canal were found (471 - 15,174 mg Sn kg^{-1} and 17 - 113 mg Pb kg^{-1}) (Suteerasak and Bhongsuwan, 2008). In support of reducing the pressure of multiple stressors on aquatic environment and for optimal management of the water resource, this study is aimed to evaluate the pollution levels of the selected metals (Pb, Sn, and Zn) in sediments at Saphan Hin, Phuket province. The information on how sources of contamination change over the years that can influence the contaminant fate in the environment then can be assessed. Pollution control measures and/or guidelines could then be developed and implemented for site management.

2. Materials and methods

2.1 Studied area and sediment collection

The study area is located at Saphan Hin, Muang District, Phuket province in the Andaman Sea. A Global Positioning System (GPS) was used to determine the coordinates of the sampling sites (Figure 1) (point A longitude 7° 51' 42.2" N; latitude 98° 24' 08.3" E and point B longitude 7° 51' 36.7" N; latitude 98° 24' 11.7" E). For sediment sampling, a stainless steel Russian corer of 60 m long (7.5 cm x 60 cm) was employed to achieve complete sediment profile collection within the sea coast at Saphan Hin area on May 2016. After sampling, the sediment samples were carefully removed from the core and were sub-sampled on site precisely by cutting into a slice according to the difference in sediment colour and texture observed visually. There are a total of 24 and 22 layer depths of the sediment samples for core A and B. All sediment samples were then immediately stored in polypropylene bags and kept in the icebox at 4 °C before taken to the laboratory at Prince of Songkla University, Phuket campus for further processing and analysis.

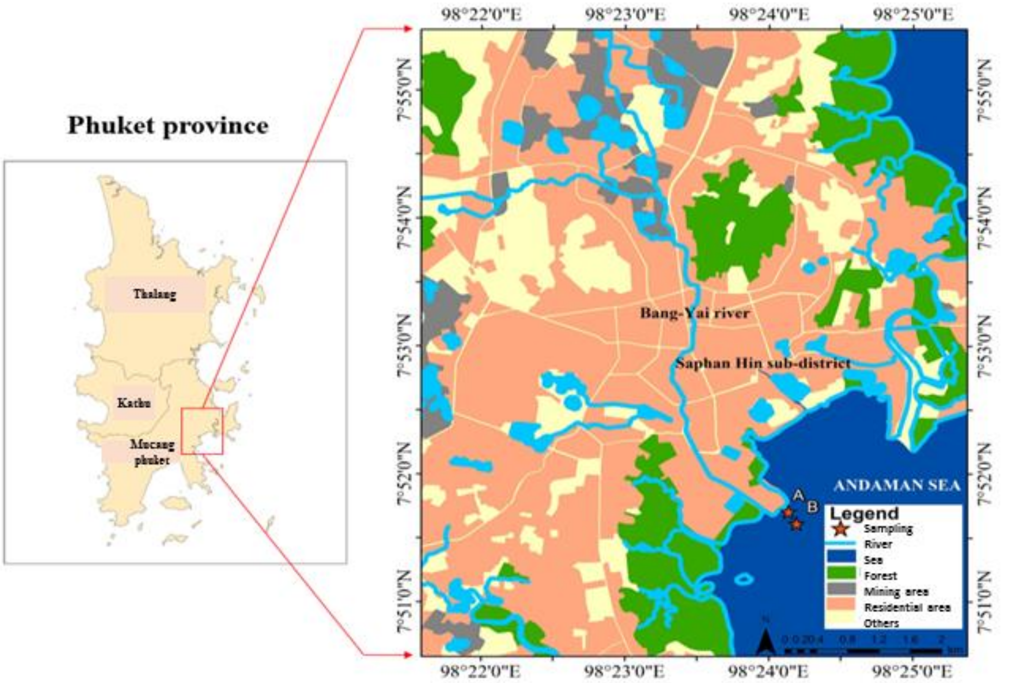

Figure 1: Sampling sites

2.2 Sediment preparation and analysis

Each slice of sediment samples was dried at 60 °C until constant weight is achieved and ground manually in a porcelain mortar. The sediment samples were then separated into two sediment fractions of fine (< 74 µm) and coarse grain particles (> 74 µm) using mesh sieve. For determination of total Pb, Sn, and Zn concentrations, aqua regia digestion according to the Standard US-EPA Method 3052 (US EPA, 1996) was performed. Extracts were analysed by ICP-OES (Perkin Elmer Optima, 4300 DV/Perkin Elmer Optima 800). For method validation, the accuracy of the analytical procedures was checked using marine sediment certified reference material (MESS-4) from the National Research Council of Canada. The results of the MESS-4, in the form of the mean values and their standard deviations, by total digestion (US EPA Method 3052) (Pb 20.75 ± 0.63 and Zn 139.87 ± 3.83 mg kg^{-1}) showed acceptable agreement with the certified values (Pb 21.5 and Zn 147 mg kg^{-1}).

2.3 Environmental risk assessment

In this study, the degree and the origin of metal pollution in sediments were assessed using the I_{geo} and the EF. The I_{geo} allows the assessment of heavy metal contamination by comparing the measured metal concentration with the metals in earth crust and is calculated as Eq(1).

$$I_{geo} = \log2 (C_n/1.5\ B_n) \qquad (1)$$

where C_n is the total metal concentration in sediments, B_n is the concentration of background value for the metal in sediments, and 1.5 is a factor for normalisation of background metal concentrations in sediments to minimise the effect of possible variations (Abrahim and Parker, 2008). In addition, the EF is calculated using Aluminium (Al) as the normalising element for crust-derived material. The EF is defined as Eq(2).

$$EF = (M_x/Al_x)_{sample}\ /\ (M_c/Al_c)_{crust} \qquad (2)$$

where $(M_x/Al_x)_{sample}$ is the ratio of metal and Al concentrations in the sediment sample, and $(M_c/Al_c)_{crust}$ is the ratio of metal and Al concentrations of the background. The background concentrations of Al, Pb, and Zn in the average earth crust of Phuket province used in this study are: 115,360 mg kg^{-1}, 20 mg kg^{-1}, and 80 mg kg^{-1} (Uduma and Jimoh, 2013).

2.4 Statistical analysis

Principal Component Analysis (PCA) and Pearson correlation analysis were used in an attempt to determine the relationships between metal concentrations, and the test variables which are known to have an influence on metal concentrations using the SPSS (2015)

3. Results and discussion

3.1 Sediment characterisation

Metal distribution in the water and sediment are dependent on a number of factors, which include pH and the metal concentrations (Balintova et al., 2012). It was found that the pH values of sediment samples in Core A and B exhibited the similar trend of slightly acidic to slightly alkaline (pH 6.8 - 8.3 and pH 6.9 - 8.1 for Core A and B) with minor fluctuation from surface to bottoms. Many studies had reported no significant variation in the pH along the cores (Zhang et al., 2014). The pH of the sediments at gulf of Thailand, on the opposite side of Andaman Sea to the right of Thailand's peninsula, varied between 4.52 and 7.76 (an average of 7.42) and showed higher values in deep areas and lower values near coastlines (Liu et al., 2016). The oxidation reduction potential (ORP) of sediment samples were measured as it is an important parameter controlling heavy metal stability and mobility (Zhang et al., 2014) and as indicator to understand relative status of the sediment (Muralidhar et al., 2016). In this study, the ORP of sediments in Core A and B ranged from -236 to -158 mV and -245 to 172 mV. This implied that the sediments in the two sampling sites were mostly in reduced form. Changes in the oxidation state of the metals associated with the oxides can greatly affect their solubility and mobility in soil and aqueous environments (Lee, 2006). As sediments become anaerobic, the redox potential decreases and transformation to the more soluble reduced forms of metals take place. Higher values of ORP were observed in the southwestern part of the upper gulf of Thailand (-388 to 312 mV, with an average of -127 mV), possibly representing the combined result of freshwater inflow and circulating currents (Liu et al., 2016).

The heavy metals entering the sea coast can be either directly dissolved in water or adsorbed to sediments (Wang et al, 2017). In this study, the overall metal concentration ranges were found as follows: 9.04 - 32.17 mg kg^{-1} and 6.33 - 36.52 mg kg^{-1} in Core A and B for Pb, and 32.83 - 68.49 mg kg^{-1} and 21.63 - 73.59 mg kg^{-1} in Core A and B for Zn (Table 1). Sn concentration was lower than the detection limit of the instrument (<0.03 mg kg^{-1}). The observed metal concentration were below the sediment quality guidelines of TEC (Pb 35.8 mg kg^{-1} and Zn 121 mg kg^{-1}) (Helen et al., 2016) and were found relatively lower than other reported studies (Thongraar et al., 2015). For example, the concentrations of Pb and Zn in the surface sediments of the western Gulf of Thailand ranged from 4.13 to 38.75 mg kg^{-1} and from 1.63 to 78.95 mg kg^{-1} (Liu et al., 2016).

Statistical analysis showed significant difference in metal concentration in different sediment layer depths ($p < 0.05$). The two elements exhibited similar concentration profile for Core A and B with an increase in metal concentrations as a function of increasing depth. The higher metal concentration in the middle and lower layer depths may be due to the long term dispersal of heavy metals in a catchment as affected by historic tin mining in the past. Sn mineral transportation was taken in the past 20 y around Saphan Hin area and this might cause the accumulation of heavy metals in the middle section of the cores. The lower metal concentration at the top layers of the sediment might be as a result of a land reclamation project for a new land used for parks and public facilities at Sapan Hin area. The dispersion of particles might cause the low metal concentration within these layers.

Table 1: Metal concentration (mg kg^{-1}) in the two studied cores at Saphan Hin area

Core A			Core B		
Depth (cm)	Pb (mg kg^{-1})	Zn (mg kg^{-1})	Depth (cm)	Pb (mg kg^{-1})	Zn (mg kg^{-1})
1	9.04 ± 0.21	32.83 ± 1.33	1	6.45 ± 0.6	21.63 ± 0.2
3	9.07 ± 0.00	39.51 ± 1.69	3	6.33 ± 0.5	22.30 ± 0.2
6	11.03 ± 0.69	41.63 ± 0.04	5	6.49 ± 1.0	22.50 ± 0.6
9	14.83 ± 0.53	50.23 ± 0.06	7	7.09 ± 0.4	25.70 ± 0.4
12	13.39 ± 0.31	44.90 ± 0.20	10	12.81 ± 0.0	38.03 ± 0.2
15	12.86 ± 0.67	48.69 ± 0.75	13	13.28 ± 0.7	41.93 ± 0.6
17	16.90 ± 1.49	44.62 ± 0.81	16	15.99 ± 1.9	49.07 ± 0.11
19	17.92 ± 0.37	53.15 ± 1.41	18	18.53 ± 0.1	57.29 ± 0.9
21	18.10 ± 0.98	50.62 ± 0.84	20	18.34 ± 0.0	57.25 ± 0.6
23	16.14 ± 0.81	56.96 ± 0.41	22	18.31 ± 1.4	49.89 ± 1.0
25	19.23 ± 0.56	54.00 ± 1.57	24	18.23 ± 1.6	43.77 ± 1.0
27	19.47 ± 0.60	58.13 ± 0.33	26	19.87 ± 1.9	46.31 ± 0.6
29	32.17 ± 1.16	55.77 ± 0.90	28	20.53 ± 0.6	50.89 ± 0.5
31	21.69 ± 1.35	56.01 ± 0.94	30	18.94 ± 0.6	43.88 ± 0.6
32	22.47 ± 0.34	53.68 ± 0.19	33	25.16 ± 0.4	60.91 ± 0.6
34	21.34 ± 0.49	59.80 ± 0.39	36	20.82 ± 1.4	52.07 ± 0.8
36	21.93 ± 0.97	55.15 ± 0.94	39	29.41 ± 1.0	64.68 ± 0.6
39	20.94 ± 0.06	59.62 ± 0.54	42	36.52 ± 0.9	73.59 ± 0.7
41	18.01 ± 0.5	45.83 ± 0.76	44	28.07 ± 0.5	69.38 ± 1.2
42	19.03 ± 0.65	68.49 ± 1.36	46	20.46 ± 0.1	53.94 ± 0.7
44	14.10 ± 0.27	54.31 ± 0.16	48	20.48 ± 0.1	48.13 ± 0.8
47	15.56 ± 0.20	44.19 ± 0.04	50	21.05 ± 0.8	56.27 ± 0.5
49	18.49 ± 0.04	49.77 ± 0.25			
51	20.28 ± 0.11	50.65 ± 0.13			

3.2 Metal risk assessment

In this study, Al was used as a reference metal in earth crust because of its high natural concentration, minimal anthropogenic contamination, and the metals to Al proportions in the crust are relatively constant (Naji and Ismail, 2011). According to the results, the I_{geo} of Pb and Zn in all sediment layer depths showed the I_{geo} values of less than zero, exhibited unpolluted of the sediments, except at the 29 cm depth layer of Core A (I_{geo} = 0.10) and 42 cm Core B (I_{geo} = 0.28) that revealed unpolluted to moderately polluted. It is suggested that nearly all the analysed sediment samples were not contaminated with the studied metals. The low I_{geo} values of the metals were found in other studies (Krishnakumar et al., 2016). For example, in the western Gulf of Thailand, on the opposite side of the Andaman sea, the I_{geo} of Pb and Zn ranged from -2.9 to 0.33 and -5.03 to 0.57 (Lui et al., 2016). Pascual-Aguilar et al.(2016) found the Igeo index ranging from low pollution to very high pollution for Zn, and for Cd with low to moderate contamination in the sediments of the River Turia, the East of the Iberian Peninsula. The EF value was used for assessing the enrichment degree of the studied metals in sediments to differentiate the sources and the results showed that the EF values of Pb in the two sediment cores (Core A and B) are between 2.0 and 6.0 indicated moderate enrichment, except at 42 cm and 50 cm depth in Core B (EF_{Pb} =1.93 and 1.88). The highest EF_{Pb} value was found in Core A at the 29 cm depth (EF_{Pb} = 6.08) showing significant enrichment. Alternatively, the EF values of Zn of the two cores were between 1.0 and 4.0. The highest EF_{Zn} values were observed in the superficial sediment at 3 cm depth for Core A (EF_{Zn} = 4.0) and Core B (EF_{Zn} = 3.8), indicating moderate enrichment. The EF values in this study (EF > 1.5) indicates a significant portion of the trace metals was delivered from non-crustal materials and that may be delivered by other sources (Barbieri M., 2016).

3.3 Correlation

The metal concentrations including Pb and Zn and sediment characteristics (pH, ORP, and EC) were used for statistical analysis to determine their interrelationships using PCA. Rotation method used was Oblimin with Kaiser Normalization. The total variance shows the significant factors and the percent of variance explained by each of variables before undergoing rotation which accounted for 83 % of the total variance. Loading values suggest a correlation between certain elements and soil characteristics of analysed data. A component plot in rotated space is given in Figure 2 which illustrates the three Principal Components (PCs). The PCA analysis results show that the first component (PC1) has the highest factor loading (5.919) followed by the second component (PC2) and the third component (PC3) with factor loadings of 5.201 and 1.439. The PC1, accounts for the most important associations, and is strongly correlated with Pb and Zn in both cores as illustrated by the high factor loading in PC1 and 57.894 % of total variance. Variables that are correlated with one another are combined into factors which are thought to be representative of the underlying correlations (Zhang, 2006). The results imply that Pb and Zn originate from the same sources hence the strong correlations and interrelationships. Pb and Zn are often found together in ore deposits (Zhang et al., 2008). The PC2 accounted for 13.438 % of total variance which dependent upon pH and ORP of the sediments, especially in Core B. Alternatively, the PC3 comprised only the EC and accounted for 11.746 % of total variance.

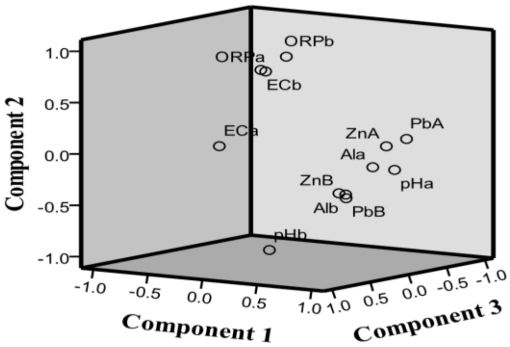

Figure 2: The three Principal Components (PCs)

4. Conclusion

The pollution levels of the selected metals (Pb, Sn, and Zn) in sediments were evaluated at Saphan Hin, Phuket province. The concentrations of Pb and Zn in the two studied cores are ranged from 9.04 - 32.17 mg Pb kg^{-1} and 6.33 - 36.52 mg Pb kg^{-1}; 32.83 - 68.49 mg Zn kg^{-1} and 21.63 - 73.59 mg Zn kg^{-1}, in Core A and B that are below the sediment quality guidelines of TEC (Pb 35.8 mg kg^{-1} and Zn 121 mg kg^{-1}). It is suggested that most of the analysed sediment samples were not being contaminated and deficiency to minimal enrichment with the studied metals due to the low I_{geo} values and the EF_{Pb} values. Metal contamination in benthic organisms and shellfish in this coastal area should be investigated for future work to evaluate the risk associated with the hazard of heavy metal accumulation in edible seafood.

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