

# SEASONAL VARIATION OF GRAIN SIZE AND HEAVY METALS CONCENTRATION IN SEDIMENT OF SUNGAI PERLIS

(Variasi Musim Saiz Zarah Dan Kepekatan Logam Berat Di Dalam Sedimen, Sungai Perlis)

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#### Abstract

Heavy metals concentration (As, Co, Cu, Pb, Zn, Cr) and grain size analysis were determined in sediment collected from Sungai Perlis during Southwest and Northeast monsoon. Ten sampling areas of the Sungai Perlis were selected based upon anthropogenic activities which resulted in elevated metals load into the river. Heavy metals concentration in the sediments show average decreasing trend of Zn>Cr>Pb>Cu>As>Co during Southwest monsoon and Cr>Zn>Pb>As>Cu>Co on Northeast monsoon which can be related by the impact of the anthropogenic activities. Most of the sampling areas were dominated by silt sediments while sand and clay sediment were poorly present. Result of grain size showed that the average mean size, sorting and skewness are  $7.32 \pm 0.44$  phi,  $1.55 \pm 0.14$  phi and  $-0.16 \pm 0.19$  phi for Southwest monsoon and  $7.14 \pm 0.27$  phi,  $1.56 \pm 0.12$  phi and  $-0.08 \pm 0.17$  phi during Northeast monsoon. The analysis of heavy metals and grain size fractions of sediments indicated that the distributions of metals and grain size in upstream and downstream river sediments were affected by monsoon seasons.

Keywords: grain size, metals, sediment, Sungai Perlis, seasonal variation

#### **Abstrak**

Kepekatan logam berat (As, Co, Cu, Pb, Zn, Cr) dan saiz zarah telah ditentukan di dalam sedimen yang diperolehi daripada kuala Sungai Perlis ketika musim monsun barat daya dan timur laut. Sepuluh kawasan telah ditentukan di kuala Sungai Perlis berdasarkan aktiviti manusia yang akan menyumbang kepada kepekatan logam berat di dalam sungai. Kepekatan logam berat di dalam sungai menunjukkan gaya penurunan Zn>Cr>Pb>Cu>As>Co ketika musim monsun barat daya dan Cr>Zn>Pb>As>Cu>Co ketika musim monsun timur laut di mana ia mungkin dipengaruhi oleh aktiviti manusia. Kebanyakan kawasan sampel diambil menunjukkan dominasi lumpur yang halus tetapi pasir jarang sekali dijumpai. Keputusan saiz zarah menunjukkan purata nilai min, susunan dan skewness adalah  $7.32 \pm 0.44$  pai,  $1.55 \pm 0.14$  pai dan  $-0.16 \pm 0.19$  pai untuk monsun barat daya dan  $7.14 \pm 0.27$  pai,  $1.56 \pm 0.12$  pai dan  $-0.08 \pm 0.17$  pai ketika musim monsun timur laut. Keputusan analisis logam berat dan saiz zarah di dalam sedimen menunjukkan taburan logam dan saiz zarah di muara sungai adalah dipengaruhi oleh musim monsun.

Kata kunci: saiz zarah, logam berat, sedimen, Sungai Perlis, variasi musim

#### Introduction

The increasing growth of human population has lead to the land development alongside of the river basin. Most of the rivers which were affected by the industrialization and urbanization become the end point where all of the effluents are discharge directly from the factories and municipal waste. This phenomenon gives stress to aquatic ecosystem which soon giving rise to water pollution and environmental deterioration. Pollutant such as heavy metals usually being carried by the runoff from the primary sources and spread to other part of the aquatic ecosystem [1].

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Elemental concentrations in sediments were not only depending by natural and anthropogenic sources but also upon the organic matter content, mineralogical composition and textural characteristic of the sediment [2]. Heavy metals are usually enriched in the smaller grain size fractions [3]. Smaller particles have a large surface area over volume ratio which is associate to co-precipitation and complexation of metals on particles surfaces. Thus, the adsorption process of metals are higher in fine grain size which result in high metals concentration [4].

There is not much data available on the heavy metals and grain size analysis in the sediment in North Western Part of Peninsular Malaysia especially in Sungai Perlis area. Contamination from anthropogenic sources such as domestic sewage, boat activities, agriculture and aquaculture run off can possess a serious threat to the food chain system in aquatic environment. Moreover, bioavailability of metals in sediment can be influenced by several factors including sediment characteristics such as grain size distribution, mineral composition and organic content [5].

Comparison of heavy metals distribution with grain size allowed the identification of the area where the sediment fine fraction appears affected by heavy metal anthropogenic contamination [6]. Therefore, it is important to study the distribution of the potentially toxic metals and its factors that can contribute to the increasing load of metals in the sediment. This research was carried out to illustrate the distribution of selected heavy metals (Co, Cu, Zn, Pb, As and Cr) and grain size distribution in Sungai Perlis during Southwest and Northeast monsoon seasons.

# **Materials and Methods**

# **Sampling Areas and Sample Collection**

Sampling was carried out in Sungai Perlis in Perlis which is located at the Northern West of Peninsular Malaysia. The location of Kuala Perlis Jetty at the edge of ocean plays an important role in attracting local people and tourist since it is one of the gateway to the Langkawi Island. This hectic river also serves as fish landing sites and anchorage for hundreds of fishing boats mainly at the upstream of river. Furthermore, there are various sources of effluent being dump straight to the river such as runoff from urban areas, agriculture and aquaculture practice and municipal waste. Generally, the river basin is approximately 310 km with the length is about 11 km which is connected from Kangar city to Kuala Perlis.

Samples were collected during Southwest and Northeast Monsoon season. Ten sampling stations were chose based upon the anthropogenic sources nearby the location. Figure 1 shows the location of the ten sampling stations and Table 1 are coordinates of each sampling station which were determined by using the Global Positioning System (GPS) device. The surface of sediments in each station was collected by using Van Veen Grab. Sediment samples were placed in clean Polypropylene (PP) plastic bags all samples were properly labeled to avoid any cross contamination.

# **Heavy Metals Analysis**

The samples were dried in oven at 80°C and grounded by using mortar and pestle. Then the samples were sieved through a 63 µm mesh size. For the analysis of total heavy metals, sediment samples were digested by following the published methods with some modifications [7, 8]. Before digestion, fine powdered samples were heat to get a constant weight. After that, a mixed acid solution (concentrated HF, HNO<sub>3</sub> and HCl) was added into the Teflon vessel along with the samples and heated at 150°C. After cooling, a mixed solution of EDTA and boric acid were added and once again the vessel was heated in the oven 150°C. After cooling, the digested samples were transferred into a clean polypropylene test tube and analysis using inductively-coupled plasma mass spectrometer (ICP-MS).

# **Grain Size Analysis**

Mean, sorting and skewness in sediment samples were determined by using laser diffraction method. Firstly, all carbonates were dissolved by using 4 M Hydrochloric acid solution (HCl). Then the organic components from sediment samples were removed by adding 50% hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>). Subsequently, the flocs of finer particles were break up by adding a dispersing agent (5% calgon solution). Then, the samples were analyzed by using Particle Size Analyzer (PSA) system. Percentage of silt, clay and sand were calculated to determine the texture of sediment using classification standard proposed by Nichols and Biggs (1985) [9].

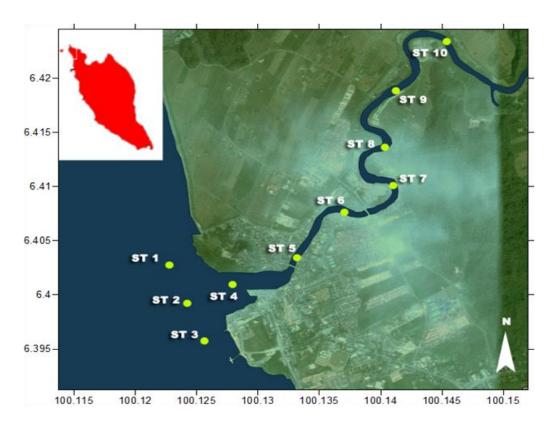


Figure 1: Sampling areas in Sungai Perlis

Table 1: The coordinates for each sampling station

Station	Latitude	Longitude		
1	6°24'27.72"N	100° 6'56.46"E		
2	6°24'2.04"N	100° 7'6.72"E		
3	6°23'42.00"N	100° 7'20.22"E		
4	6°24'1.62"N	100° 7'34.92"E		
5	6°24'10.68"N	100° 7'58.44"E		
6	6°24'26.22"N	100° 8'15.30"E		
7	6°24'38.10"N	100° 8'22.02"E		
8	6°24'53.70"N	100° 8'22.32"E		
9	6°25'7.20"N	100° 8'27.72"E		
10	6°25'26.10"N	100° 8'34.08"E		

#### **Results and Discussion**

# **Sediment Grain Size**

Characteristics of sediment such as mean size, sorting and skewness are given in Table 2. Current and seasonal changes tend to affect the distribution of sediment characteristics in Sungai Perlis. The sampling points extend from downstream to upstream river which cover approximately 7 km of the river. Since the study areas were affected by several human activities such as boating and land reclamation, sediment composition and grain size may fluctuate overtime. Thus, grain size analysis may indicate the impact of different activities to the sediment in the aquatic environment [11].

Stations	Southwest Monsoon			Northeast Monsoon			
	Mean	Sorting	Skewness	Mean	Sorting	Skewness	
1	7.04	1.79	0.02	6.70	1.77	-0.07	
2	7.27	1.59	-0.29	6.84	1.63	-0.04	
3	8.50	1.37	-0.43	6.94	1.57	0.05	
4	7.16	1.56	-0.21	6.99	1.53	-0.04	
5	7.27	1.54	-0.37	7.18	1.54	-0.07	
6	7.32	1.53	-0.19	7.48	1.31	0.08	
7	7.27	1.43	0.02	7.29	1.57	-0.39	
8	7.21	1.45	-0.14	7.14	1.69	-0.37	
9	7.34	1.78	-0.19	7.39	1.46	0.02	
10	6.86	1.45	0.19	7.48	1.52	0.00	
Average	7.32	1.55	-0.16	7.14	1.56	-0.08	
Std Dev	0.44	0.14	0.19	0.27	0.12	0.17	
Classification	Fine	Poorly	Symmetrical	Fine	Poorly	Symmetric	
	silt	soorted		Silt	sorted		

Table 2: Characteristics of sediments during Southwest and Northeast Monsoon seasons

The average mean size of sediment along Sungai Perlis during Southwest (SW) and Northeast (NE) monsoons was  $7.32 \pm 0.44$  phi and  $7.14 \pm 0.27$  phi, classified as fine silt for both season. The dominating of high percentage of fine silt may result from the low velocity depositional in the river [12]. Apparently, the mean size reach a maximum value of 8.50 phi (Station 3) and minimum value at Station 10 (6.86 phi) during SW monsoon. However, during NE monsoon, maximum value was obtained at Station 6 and Station 10 (7.48 phi) and the minimum value is 6.70 phi at Station 1. SW monsoon which characterized by strong wind mainly affect the western coastal which can be explained by the difference on the mean size on both season. Generally the mean size gives a simple indication of the magnitude of the force, applied by water or wind which will affect the mean size of the grains [13].

Most of sampling sites during these two seasons were poorly sorted which also indicate that little selection of grains size had taken place during transport or deposition. The fluctuation of sorting values on each sampling sites reflected the unstable condition of the environment in the estuary which may be due to human activities and natural constituent [6].

The skewness of Sungai Perlis sediment reach a maximum value of 0.19 phi at Station 10 during SW monsoon and a minimum value at Station 3 (-0.43 phi). About 60% of the samples were negatively skewed (i.e. coarsely skewed), while 40% of the sediments were positively skewed (i.e. finely skewed) during NE monsoon. Based on Table 2, the average skewness during NE monsoon show negatively skewed data compare to SW monsoon but still this two seasons indicate symmetrical skewness which could probably be associated with sediments erosion in environments dominated by strong current actions. Some of sampling sites on both seasons show tendency for sediment to become coarser, due to increasing transport capacity and current within the river [12].

Accordingly, SW monsoon had finely silt compare to NE monsoon which explain the role of the wind on the distribution patterns of sediments in the study area. Besides, influence from natural activities (i.e. transport capacity and current) and human activities (boating and land reclaimation) around the study area might be effected the distribution pattern of sediment in the study area.

# **Heavy Metals Analysis**

The concentrations of heavy metals in sediment of Sungai Perlis estuary during Southwest (SW) and Northeast (NE) monsoon are shown in Table 2. Heavy metals concentration in the sediments show average decreasing trend of Zn>Cr>Pb>Cu>As>Co during SW monsoon and Pb>Cr>Zn>Cu>As>Co on NE monsoon season. The average concentration of Cr was slightly higher during NE monsoon compare to SW monsoon. The maximum value of Cr was found at Station 8 (158.23 mg/kg) and the minimum value was at 70.92 mg/kg (Station 3) during NE monsoon. Primarily Cr was used in paints especially on boat's hull in order to slow down the growth of organisms that attach to the hull [3]. The dumping of municipal and industrial waste materials can significantly increase chromium concentration in soil and is usually accompanied by groundwater contamination [15]. Since SW monsoon bring a heavy rainfall in the Perlis, Cr concentration was diluted which explained the slightly lower concentration compare to NE monsoon.

The average concentration of Zn (94.45 mg/kg) obtained was slightly higher during SW monsoon compare to NE monsoon that only has a value of 90.49 mg/kg. Based on Table 2, concentration of Zn reach a maximum value at Station 10 (121.00 mg/kg) and minimum value of 62.20 mg/kg at Station 4 during SW monsoon. During SW monsoon, a slightly high humidity affect the weather condition and approximately 35%-110% above average of rain was received in Perlis [18]. During this rainy season, runoff from the industrial areas and the main city of Perlis (Kangar) were carried to the Sungai Perlis which can be explained the elevated level of Zn in the river. This result was in the line with the finding by Fortune (2006) where storm water from urban areas has increase the Zn loads to 60 times higher than background levels [12]. Besides, other sources of Zn alongside the Sungai Perlis include residential and domestic waste may also contribute to the load of Zn in the river. The calculation for household products includes laundry detergents, general purpose cleaners, shampoos, bar soaps and toilet tissue could account for a portion of Zn load from these sources [16]. Furthermore, lower salinity in the estuary river due to the rainfall might also affect the mobility and availability of metals the sediment. According to Laing et al., (2009) the binding of Zn in the sediment was affected by salinity where decrease in salinity might increase the mobility of Zn in the river [17]. On the other hand, Perlis received a minimum rainfall which is below average level during NE monsoon season [18]. The difference in between these monsoons might explain the high level of Zn in the sediment of Sungai Perlis estuary.

In contrast, average concentrations of Pb (52.30 mg/kg) during NE monsoon was slightly higher compare to SW monsoon with the value of 39.23 mg/kg. The maximum value of Pb was obtained at Station 9 which has a value 76.88 mg/kg during NE monsoon and the minimum value of 32.46 mg/kg at Station 3. Elevated concentrations were found at a number of sites mostly at downstream areas. Due to the location of Perlis at the west coast of Peninsular Malaysia, NE monsoon do not really affect the western coastal area [18] which, by this time, high number of fishing and boating activities were spotted compare to SW monsoon season. Oil leakage from boats may be contributed to the Pb source which originates from leaded fuel use by fishing boat [12]. Besides, due to the fact that Sungai Perlis is the main river that carry waste from urban and industrial areas, there is no doubt that Pb was accumulated in the sediment. Unsal (2001) reported that local industries can cause lead pollution in the aquatic environment and a notable amount of lead has been transported in particulate form contained in riverbed [21].

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Table 3: Heavy metals concentration (mg/kg) in the sediment during Southwest and Northeast Monsoon

Stations	Monsoon	Heavy Metals Concentration (mg/kg)						
		Cr	Zn	Pb	As	Cu	Co	
1	SW	77.01±2.23	84.12±2.84	33.34±4.51	11.88±1.25	34.47±4.46	6.01±0.47	
	NE	76.73±3.02	62.82±1.25	39.71±1.70	15.52±1.12	$10.22 \pm 0.25$	$6.78 \pm 0.88$	
2	SW	82.34±1.54	73.99±4.33	31.19±2.15	13.98±2.73	14.49±6.02	6.69±0.95	
	NE	105.27±5.14	86.16±2.31	46.83±1.69	23.65±1.79	16.76±1.77	10.26±1.35	
3	SW	90.85±4.02	89.08±6.34	38.42±4.21	18.98±1.32	19.74±2.36	8.18±2.00	
	NE	70.92±2.98	61.29±2.33	32.46±1.21	26.48±1.68	7.32±1.49	8.12±1.96	
4	SW	56.67±3.17	62.20±2.68	23.54±4.73	11.47±0.96	8.01±5.51	4.74±0.43	
	NE	114.47±4.69	95.33±4.41	52.16±4.00	32.87±3.37	18.13±4.44	11.97±1.85	
5	SW	113.17±5.65	107.63±6.66	45.87±1.70	20.19±3.59	22.74±3.09	9.19±2.72	
	NE	103.54±3.04	78.48±2.75	43.47±1.36	26.41±3.75	15.16±2.20	9.80±1.04	
6	SW	100.76±3.11	109.69±6.86	45.62±2.96	24.98±3.44	29.03±4.62	10.75±1.70	
	NE	115.34±5.30	89.53±4.92	48.32±0.56	29.59±2.46	23.34±4.21	$10.73\pm1.02$	
7	SW	109.38±2.98	111.04±3.48	45.97±1.84	26.36±4.42	30.57±6.60	10.70±1.67	
	NE	$124.44\pm6.08$	96.64±1.46	48.57±6.24	26.27±0.11	$28.53\pm2.37$	10.58±1.59	
8	SW	96.52±6.11	100.34±4.40	42.66±3.86	21.47±0.97	21.43±5.35	9.74±0.99	
	NE	158.23±7.23	117.37±.40.09	63.99±2.72	35.72±1.85	20.73±3.52	14.41±4.11	
9	SW	87.52±5.20	85.35±3.70	36.19±3.00	19.94±0.12	19.86±4.26	7.50±0.59	
	NE	126.75±3.15	110.03±1.90	76.88±4.43	77.50±7.02	$26.4\pm2.46$	24.17±6.91	
10	SW	116.64±4.58	121.10±4.09	49.44±1.15	28.13±1.43	35.91±2.56	11.70±2.38	
	NE	132.60±2.55	$107.30\pm3.02$	70.62±1.73	59.12±2.13	35.29±3.14	$17.89\pm2.51$	
Average	SW	93.09	94.45	39.23	19.74	23.63	8.52	
	NE	112.83	90.49	52.3	35.31	20.19	12.47	
Std	SW	18.35	18.57	8.18	5.86	8.87	2.28	
Dev	NE	25.81	18.95	14.01	18.72	8.51	5.17	

It was found that concentration of As was slightly higher during NE monsoon (35.31 mg/kg) compare to SW monsoon (19.74 mg/kg). The sources of As was possibly byproduct from agriculture and aquaculture activities that uses As in pesticides and as additives in fish/shrimp food [22]. Furthermore, Cu concentration shows a maximum value at Station 10 (35.91 mg/kg) during SW monsoon and a minimum value of 7.32 mg/kg at Station 3 during NE monsoon season. Municipal waste and boating activities alongside the river may increase Cu load since the major source of Cu were from vehicles (brakes, tires, protective paint on boats, petrol, car washing) and building materials such as drinking water pipes and corrosion from steel material [23]. Sometimes, a lot of rubbish can be seen visible at the river such as plastic, tires, and even corroded steel which being lift during grabbing the sediment. On the other hand, Co concentration shows a maximum value at St 10 (11.70 mg/kg) and a minimum value of 4.74 mg/kg during SW monsoon. Improper sewage from squatter and residential areas alongside the river may possibly contribute to the Co load in the sediment [24].

# Conclusion

From the data obtained, mean size and skewness during SW monsoon shows slightly higher compare to NE monsoon season and for sorting is vice versa. However, classification of mean, sorting and skewness were the same for both monsoon seasons. For heavy metals, Cr concentration was lower during SW monsoon which bring a heavy rainfall in the Perlis and cause Cr concentration diluted. On the other hand, rainy season during SW monsoon tend to carry runoff from the industrial areas and the main city of Perlis (Kangar) and elevate the level of Zn in the river. The old dumping sites, aquaculture, anchorage and residential areas were the possible source of various metals to the river. The main reason behind the seasonal variation of heavy metals is dilution of metal ions in the sediment during rainy season and it tends to accumulate free movement of the metal ion in the next seasons.

# Acknowledgement

This research was conducted with funding from the FRGS research grant. The author thank to the Centre of Ocean Research, Conservation and Advances (ORCA) teams for their invaluable assistance throughout the sampling period.

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