**ORIGINAL ARTICLE** 



# Heavy metals pollution sources of the surface water of the Tunggak and Balok river in the Gebeng industrial area, Pahang, Malaysia

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#### Abstract

The heavy metal contamination of surface water is regarded as one of the most serious environmental hazards. The purpose of this study is to identify the origins of 10 heavy metals found in surface water in the Gebeng industrial area rivers. The samples were gathered over the course of a year at ten stations along the Tunggak and Balok rivers. According to the Malaysian heavy metals standard, the surface water of the Gebeng rivers was contaminated with six heavy metals: Cd, Co, Cu, Pb, Mn, and Ni. Inductively Coupled Plasma Mass Spectrometry was used to analyze the data. SPSS version 22.0, a multivariate statistical tool, was used to identify the sources. The heavy metals have been divided into three components using principal component analysis, indicating that the pollution is due to anthropogenic causes. The stations were grouped into three groups using cluster analysis, with high pollution loading falling under industrial zone stations (IZ1, IZ2, and IZ3), indicating pollution from industrial sources. In general, the findings of this research will be useful in future studies aimed to reduce heavy metal contamination in the Gebeng rivers.

Keywords Heavy metals · Gebeng industrial area · Contamination · Principal component analysis · Cluster analysis

### Introduction

The continued manufacturing of countries has led to extensive environmental problems. A broad variety of chemicals has been discovered in air, water, and soil. Water is necessary for the survival of all existing organisms. The quality of water is essential for humanity, since it is connected with human health. The natural substances which lead to water pollution are minerals, gasses, humus material, waste created by animals and other living organisms in the water. Another significant source is anthropogenic input from mining, national and industrial activities, such as discharge from wastewater from electroplating smelting, corrosion of copper tubing, and metal engraving industries. Nowadays, large quantities of untreated industrial sewage have been discharged into surface water bodies for disposal (Shazia et al., 2013).

M. S. Islam sujaul@ump.edu.my On the other hand, rapid economic and industrial development has led to the accumulation of heavy metal elements of impacted sites across the world (Hu et al., 2019). Heavy metals generally have persistent bioavailability, long residence times (commonly exceeding decades), and often low concentration thresholds indicate toxicity (Jiang et al., 2017). The excessive accumulation of heavy metals can hence disrupt the usual biochemical processes which occur in soils, leading to the deterioration of soil quality, reduced agricultural productivity and quality, and human health risks (Marchant et al., 2017).

Several studies had shown that anthropogenic activities contribute considerably to the concentration increase of heavy metal concentration in surface water above natural levels (Liu et al., 2018). In fact, heavy metals are considered the main causes of pollution in aquatic ecosystems, and their toxicity, persistence, ecological danger, bioaccumulation, and non-biodegradable nature have raised widespread concern across the world (Zhao et al., 2018). The abundance of heavy metals in the environment is mostly due to natural and human activity. Anthropogenic acts, on the other hand, can create heavy metals in water and sediment, polluting the aquatic environment (Ali et al., 2016). Because of the effects of urbanization and industrialization, heavy metal

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contamination in aquatic environments has risen (Grigoratos et al., 2014). Heavy metal contamination has risen in aquatic ecosystems in recent years as a result of fast economic and industrial growth, and it is now one of the most serious environmental issues (Monferran et al., 2016). As a result, monitoring heavy metal contaminants in water and sediment is critical for any contaminated riverine environment (Saleem et al., 2015; Sundararajan et al., 2017).

The identification of the sources of the following pollutants in the industrial environment has been documented using a variety of approaches. Principal component analysis (PCA), cluster analyses (CA), and chemical mass balance (CMB) are the most used techniques (Shi et al., 2011; Yatkin & Bayram, 2008). PCA is a method for reducing a raw data set into some principal components, and CA is used to make groups or clusters of the monitoring station, where the contaminant sources are recognized accordingly. PCA and CA were used in several prior study investigations to categorize heavy metal sources in the industrial environment (Hong et al., 2018; Mummullage et al., 2016). Because these two techniques do not include all of the raw data set's information, the number of sources may be underestimated (Zhang et al., 2012). The CMB technique is mostly used to locate pollution sources in the atmosphere (Chow & Watson, 2002).

Gebeng is a rapidly developing industrial district in Kuantan, Pahang, Malaysia, where the food and beverage, paper, chemical, textiles, palm oil, and rubber processing sectors are the most significant sources of water pollution (Sujaul et al., 2015). Because of the rapid expansion of industries, the pollution level in the nearby Gebeng watershed has increased, and most of the wastewater released from the sectors contained contaminants and was dumped into the surface water, particularly in the area of the Tunggak and Balok rivers, which are the two most important rivers in Gebeng (Salah et al., 2017; Sujaul et al., 2013). As a result, the focus of this study is on determining the origins of heavy metal concentrations in the surface water of the Tunggak and Balok rivers in the Gebeng industrial region of Pahang, Malaysia.

### **Materials and methods**

#### Study area and sampling stations

The study area, Gebeng Industrial Estate, of this investigation is one of the potential industrial regions of Malaysia. The industrial park is located between the coordinates of 03°58'00" North and 103°26'00" East. Gebeng is around 20 km from Kuantan city and close to the Kuantan seaport. The Balok and Tungguk rivers flow through this industrial region, eventually ending into the South China Sea (Salah et al., 2018; Sujaul et al., 2013). On the Tunggak and Balok rivers, ten stations were established: seven on the Tunggak River, namely, upstream (UP), industrial zone (IZ) with three stations, housing area (HA) with two stations, and downstream (DS), and the final three on the Balok River, namely, BS1, BS2, and BS3 (Fig. 1). The names, geographical coordinates, and positions of the stations are shown in Tables 1 and 2.

#### Sampling collection frequencies

The water samples were taken six times a year, from October 2016 to August 2017, three times during the wet season and three times during the dry season. Samples were collected 10 cm below from the surface water using 500 ml HDPE bottles and kept in an icebox (<4 °C) immediately before being transported to the laboratory for analysis. A number of ten (10) heavy metals: As, Ba, Cd, Co, Cr, Cu, Pb, Mn, Ni, and Zn were analyzed in the laboratory using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). All analyses were performed within seven (7) days of sampling. All chemical testing and analyses were carried out in the Environment Laboratory of the University Malaysia Pahang (UMP).

### Water sampling method

The method for water sampling was adopted from the "Standard Methods for the examination of water and wastewater" (APHA, 2005); Water analysis guide (HACH, 2005), and GEMS/Water operation guide (WHO, 2007). Water samples were collected manually from each sampling station. The following procedures were maintained during water sampling:

- I. Water samples were collected close to or in the middle of the river, wherever possible.
- II. Samples were taken approximately 10 cm from below the surface water.
- III. Sampling bottles were cleaned with chromic acid and rinsed with distilled water before the samples collection, and all bottles were rinsed again with river water before carrying out the sampling.

#### **Analytical method**

A total of ten heavy metals were analyzed in the laboratory from the collected water samples. The methods and equipment used in the analysis are shown in Table 3.

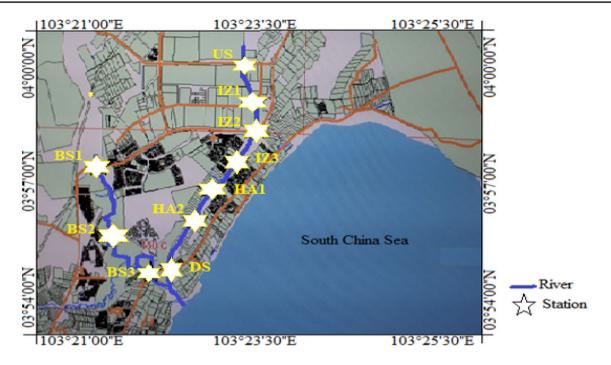


Fig. 1 Map of water sampling stations at Gebeng

Table 1 Location of the	
sampling stations and their	
geographical coordinates	

Station Number	Name of stations	Geographical coordinates	Location	
1	Upstream station (US)	N 03°59′13.8″ E 103°23′17.9″	Jalan Gebeng 2/5	
2	Industrial Zone1 (IZ1)	N 03°58'32.9″ E 103°23'18.2″	Jalan Gebeng 1/11	
3	Industrial Zone2 (IZ2)	N 03°58'12.0" E 103°23'22.2"	Jalan Gebeng 1/6	
4	Industrial Zone3 (IZ3)	N 03°57′54.1″ E 103°23′21.4″	Jalan Pintasan Kuantan	
5	Housing Area1 (HA1)	N 03°57'41.3″ E 103°23'13.7″	Jalan Balok Makmeir	
6	Housing Area2 (HA2)	N 03°57′28.6″ E 103°23′06.7″	Jalan Sebeirang Balok 3	
7	Downstream station (DS)	N 03°56'34.7" E 103°22'30.5"	Jalan Kuantan Kemanan	
8	Balok Station 1 (BS1)	N 03°59′34.8 E 103°21′27.5″	Jalan Gebeng 2/6	
9	Balok Station 2 (BS2)	N 03°57′33.3″ E103°21′47.9″	Jalan Pintasan Kuantan	
10	Balok Station 3 (BS3)	N 03°56′30.9″ E 103°22′19.3″	Jalan Kuantan Kemanan	

### **Statistical methods**

The heavy metal means concentrations and standard deviations were determined to know the metals in surface water. To examine statistical connections and identify possible sources of heavy metal elements, principal component analysis (PCA) and cluster analysis (CA) were used. SPSS 22.0 (IBM SPSS Statistics, IBM Corp., USA) and R (Version 3.4.0) were used to analyze the data in this study (Dai et al., 2018).

Parameters	Methods of analysis	Equipment/Instrument
Arsenic	EPA Method 6020A (EPA, 2007)	Agilent 750cx ICP-MS (Inductively coupled plasma mass spectrometry)
Barium		
Cadmium		
Cobalt		
Chromium		
Copper		
Lead		
Nickel		
Zinc		
Manganese		

Table 2 Methods and equipment used in analyzing the heavy metals in water

Table 3 Malaysian standard, Mean and standard deviation (SD) of heavy metals in water quality for all the stations

Stations	As	Ва	Cd	Cr	Со	Cu	Pb	Mn	Ni	Zn
Malaysian Standard	0.05 ppm	1 ppm	0.01 ppm	0.05 ppm	_	0.02 ppm	0.05 ppm	0.1 ppm	0.05 ppm	5 ppm
US	0.0041	0.2307	0.0197	0.0122	0.0781	0.0046	0.1046	0.5398	0.0414	0.7521
IZ1	0.0051	0.0542	0.0877	0.0246	0.2931	0.2444	0.0859	0.4844	0.0752	0.5339
IZ2	0.0059	0.2267	0.0171	0.0113	0.4085	0.0241	0.0881	2.1983	0.0672	0.8527
IZ3	0.0088	0.2564	0.165	0.0695	0.4874	0.3148	0.1553	0.3086	0.2273	0.5809
HA1	0.0065	0.0499	0.1602	0.0098	0.0452	0.0237	0.1168	0.4023	0.0687	0.4148
HA2	0.0067	0.2063	0.1375	0.0216	0.0458	0.0213	0.121	0.3004	0.0584	0.0827
DS	0.0092	0.1521	0.0504	0.0453	0.0466	0.1956	0.1298	0.3484	0.0559	0.2686
BS1	0.0032	0.1994	0.0036	0.0191	0.0035	0.0248	0.1014	1.3624	0.0437	0.4466
BS2	0.0038	0.0592	0.005	0.015	0.0034	0.1525	0.0954	0.3203	0.0344	0.8992
BS3	0.0041	0.0539	0.0061	0.0231	0.0035	0.0218	0.1332	0.2529	0.0449	1.1378
Mean	0.0057	0.1489	0.0652	0.0252	0.1415	0.1028	0.1132	0.6518	0.0717	0.5969
SD	0.0021	0.0856	0.0669	0.0186	0.1833	0.1142	0.0222	0.6326	0.0563	0.3172

# Principal Component Analysis (PCA)

Environmental Impact Assessment (EIA) research, such as describing EIA reports, environmental characterization of alternative corridors, and environmental characterization of local districts, have all been employed with PCA. Two PCs were discovered from 10 to 15 variables in these three investigations, and PCs were utilized as characterization criteria. The scores of alternatives were compared to the assessment criteria of three projects using PCA. PCA is a method for extracting a smaller number of uncorrelated variables, referred to as "principal components (PCs)," from a large set of data with minimal data loss and no correlations between assessment criteria.

# **Cluster analysis**

Cluster analysis (CA) is a multivariate approach for sorting a large data collection and grouping related observations (objects) into a single cluster. The observation is comparable, yet the cluster is distinct from other clusters. The total number of clusters, as well as the total number of observations in each cluster, are unknown. To do cluster analysis and reflect the data structure, a variety of methods are available. Hierarchical clustering and non-hierarchical clustering are the two techniques discovered. Hierarchical cluster analysis (HCA) will be utilized to group or cluster monitoring stations and metrics in this research.

# **Results and discussion**

# Assessment of heavy metals in surface water

The mean concentration of As was  $0.0057 \pm 0.0021$  ppm ranged from 0.0092 ppm at the IZ3 station to 0.0032 ppm at the BS1 station. The mean concentration of Ba was  $0.1489 \pm 0.0856$  ppm, where the maximum concentration was 0.2564 ppm recorded at the IZ3 station and the minimum 0.0499 ppm at HA1 station. The mean concentrations

of As and Ba were below the Malaysian threshold standard. The average values of Zn ranged from 0.0827 ppm at HA2 station to 1.1378 ppm at BS3 station with a mean value of  $0.5969 \pm 0.3172$  ppm, while the average concentration value of Cr was varied between 0.0098 ppm at HA1 and 0.0695 ppm at IZ3 station with a mean value of  $0.0252 \pm 0.0186$  ppm. Similar to As and Ba, Zn and Cr were below the Malaysian threshold level at most of the sites. The concentration of heavy metals which were measured below the Malaysian threshold level is presented in Fig. 2.

The concentration of heavy metals which were measured higher than the Malaysian threshold level is presented in Fig. 3. In the case of Cd, the highest value was estimated at 0.1650 ppm at the IZ3 station, while the lowest 0.0036 ppm was at the BS1 station with the mean value  $0.0652 \pm 0.0669$  ppm for all stations. The mean

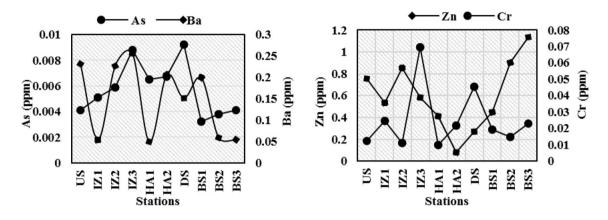


Fig. 2 Concentration of heavy metals (As, Ba, Zn, Cr) lower than the Malaysian threshold level at different sampling stations

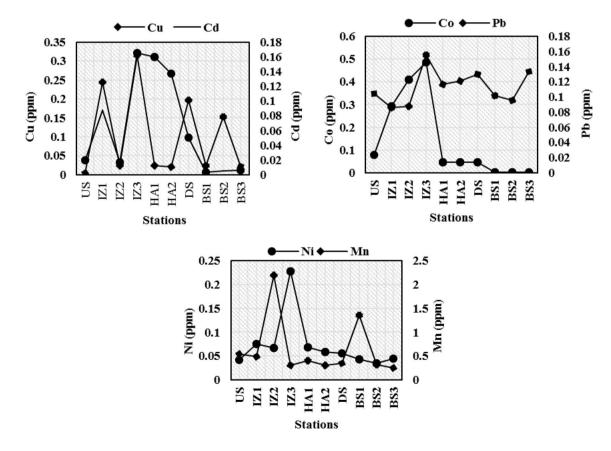


Fig. 3 Concentration of heavy metals (Cu, Cd, Co, Pb, Ni, Mn) higher than the Malaysian threshold level at different sampling stations

concentration value of Cu was  $0.1028 \pm 0.1142$  ppm. The higher value recorded 0.3148 ppm at station IZ3 and lowered value 0.0046 ppm at US station. As can be seen, the mean concentration value for Cd, and Cu exceed the Malaysian threshold limit especially at the IZ3 station, which is due to effluents from the industrial and sewage treatment plants of the Gebeng industrial area, and the obtained results are investigated by (Sobahan et al., 2013). In Malaysia, the threshold level of Co has not been recommended. Therefore, the Water Protection Section of British Columbia, Canada (2013) indicated that the threshold level of Co in the surface water is 0.0040 ppm. In this study, the average concentration of Co was  $0.1415 \pm 0.1833$  ppm, which is beyond the Canadian threshold level. The mean concentration of Pb was  $0.1132 \pm 0.0222$  ppm which was also more than the Malaysian threshold level. Similarly, Ni's mean concentration value was  $0.0717 \pm 0.0563$  ppm which eventually exceeds the Malaysian threshold limit. The highest value (0.2273 ppm) was recorded at the IZ3 station which might be due to the industries that use nickel and nickel compounds (Hossain et al., 2013), whereas the lowest value 0.0344 ppm was at the BS2 station. Finally, the mean value of Mn exceeds the threshold level of the Malaysian water at all the stations. The results of heavy metals concentration of the Tunggak and Balok rivers are presented in Table 3.

#### Principal component analysis (PCA) of heavy metals

The component plot was used to gather data for the principal component analysis to better identify and understand the sources of heavy metals. We were able to confirm the main possible pollution sources in the Gebeng region river as a result of this. Table 4 and Fig. 4 show the PCA findings of heavy metals in Gebeng surface water. The results revealed three main components with eigenvalues > 1 that accounted for 80.07% of the data matrix's total variance. As, Cd, Cr, Cu, Pb, and Ni were highly loaded (> 0.70) in the major

Table 4 Rotated component matrix of heavy metals in water

Parameters	Component						
	1	2	3				
As	0.846	-0.061	-0.248				
Ba	0.356	0.626	-0.444				
Cd	0.735	-0.259	-0.322				
Co	0.591	0.7	0.19				
Cr	0.912	0.01	0.214				
Cu	0.736	-0.006	0.448				
pb	0.728	-0.324	0.052				
Ni	0.904	0.259	0.18				
Zn	-0.393	0.267	0.792				
Mn	-0.342	0.855	-0.227				

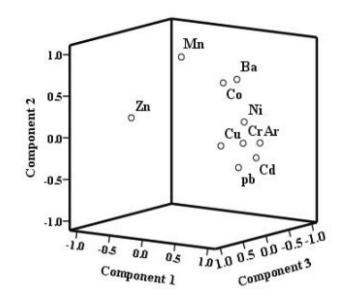


Fig. 4 Component plot of heavy metals in water

principal component (PC1), which accounted for 47.22% of the total variance. The next main component (PC2), which was heavily weighted by Co and Mn, offered 19.28% of the total variance. Only Zn is significantly loaded in the last principal component (PC3), which explains 13.58% of the total variance. Paulina et al. (2018) and Von Sperling (2017) suggested that these elements might be detected in surface water as a result of numerous human activities.

#### Cluster analysis (CA) of heavy metals

A hierarchical cluster was built to show the results of a cluster analysis of ten stations based on pollution levels (Fig. 5). The hierarchical cluster shows grouping based on heavy metal concentrations in the water at each station. The stations were divided into three groups by the CA. Cluster 1 was made up of the most contaminated stations, IZ3 and

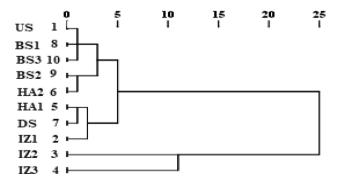


Fig. 5 Hierarchical cluster analysis of 10 stations for water heavy metals

IZ2, while cluster 2 was made up of moderately polluted stations, IZ1, DS, and HA1. Finally, low-polluted stations HA2, BS2, BS3, BS1, and the US were grouped in cluster 3. All variables were divided into four groups in the hierarchical cluster, which indicates grouping according to surface water sample locations. Cluster 1 had two sample sites (4 and 3), while Cluster 2 had one (2,7 and 5). The majority of these sites are in the Tunggak river's industrial region. Cluster 3 includes two locations (6 and 9) that are mostly found in the medals of the two rivers. The sites (10, 8, and 1) in Cluster 4 were upstream and downstream of the Balok river, and upstream and downstream of the Tunggak river, respectively. These findings show that there was substantial pollution in the industrial region. The heavy metal pollution in the Gebeng industrial region may be due to anthropogenic activity in the industrial sector.

# Conclusion

The discharge of industrial wastewater to the surface water of Gebeng is a major threat to its water quality. Several 10 heavy metals water quality parameters were measured. Ten hazardous heavy metals were found in the rivers of Gebeng, including As, Ba, Cd, Co, Cr, Cu, Pb, Mn, Ni, and Zn. However, most of the concentration of the parameter was exceeded the NWQS Malaysia recommended threshold level, especially in the industrial zone stations (IZ1, IZ2, and IZ3). The pollution source identified in the study indicated that the point sources such as the industrial effluents that contributed to the major pollutant were associated with the urban and domestic wastewater. The physicochemical parameters sources were natural, associated with some anthropogenic activities including deforestation, hill cutting, and refilling for industrial expansion. Similarly, the sources of the heavy metals were the industries effluents that used metal or metal alloys besides the agricultural run-off, fumes from vehicle exhausts, road dust associated with natural sources also contribute to the heavy metals contamination.

In particular, the water of the Gebeng River has been subjected to anthropogenic activity from the companies that have set up shop there. The water quality of the Tunggak and Balok Rivers was found to be contaminated by six heavy metals, including Cd, Co, Cu, Pb, Mn, and Ni, according to Malaysian standards, particularly at the industrial zone station (IZ3) on the Tunggak River. Heavy metals were found to come from anthropogenic sources, including industrial sources, according to PCA. The industrial zone stations IZ3, IZ2, and IZ1 were identified by CA as a high pollution loading source. Finally, cluster analysis indicated that the industrial zone and downstream stations were more polluted than others. For this reason, sustainable management approaches are required for the protection of the river water of the Gebeng area from industrial pollutants.

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#### Declarations

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper. All listed authors are qualified for authorship as authors and mutually agree to submit it for consideration. Each author has participated and contributed sufficiently to take responsibility for every portion of the content.

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