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Evaluation of Dissolved Heavy Metals in Water of the Sungai Semenyih (Peninsular Malaysia) using Environmetric Methods

(Penilaian Logam Berat Terlarut dalam Air Sungai Semenyih (Semenanjung Malaysia)
Menggunakan Kaedah Environmetrik)

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ABSTRACT

The study to determine the concentrations of dissolved heavy metals in the Sungai Semenyih and to use the environmetric methods to evaluate the influence of different pollution sources on heavy metals concentrations was carried out. Cluster analysis (CA) classified 8 sampling stations into two clusters based on the similarity of sampling stations characteristics, cluster 1 included stations 1, 2, 3 and 4 (low pollution area), whereas cluster 2 comprised of stations 5, 6, 7 and 8 (high pollution area). Principal component analysis (PCA) of the two datasets yield two factors for low pollution area and three factors for the high pollution area at Eigenvalues >1, representing 92.544% and 100% of the total variance in each heavy metals data sets and allowed to gather selected heavy metals based on the anthropogenic and lithologic sources of contamination.

Keywords: Dissolved heavy metals; environmetric methods; Sungai Semenyih

ABSTRAK

Kajian untuk menentukan kepekatan logam berat terlarut di Sungai Semenyih dengan menggunakan kaedah environmetrik untuk menilai pengaruh punca pencemaran yang berbeza pada kepekatan logam berat telah dijalankan. Analisis kelompok (AK) mengelaskan 8 stesen persampelan kepada dua kelompok berdasarkan persamaan ciri stesen persampelan, kelompok 1 terdiri daripada stesen 1, 2, 3 dan 4 (kawasan pencemaran rendah), manakala kelompok 2 terdiri daripada stesen 5, 6, 7 dan 8 (kawasan pencemaran tinggi). Analisis komponen utama (PCA) kedua-dua set data menghasilkan dua faktor bagi kawasan pencemaran yang rendah dan tiga faktor bagi kawasan pencemaran yang tinggi pada nilai Eigen > 1, iaitu 92,544% dan 100% daripada jumlah varians pada setiap set data logam berat dan membolehkan pengumpulan logam berat yang dipilih berdasarkan sumber antropogenik dan sumber pencemaran litologi.

Kata kunci: Kaedah environmetrik; logam berat terlarut; Sungai Semenyih

INTRODUCTION

In the last few decades, the rapid industrial development has added enormous loads of pollutants to our water resources such as rivers, lakes and streams. Out of these pollutants, heavy metals are of main concern due to their bio-accumulative and persistent nature (Kaushik et al. 2009). Heavy metals are natural elements found at various background levels at different places throughout the world. Heavy metals are persistent and cannot be depleted from the environment. Thus, problem arises when their availability is high due to high background levels or human activities. Heavy metals are enriched in the environment by different human activities (Srivastava et al. 2008). The results of these activities end up in outlets and wastes where heavy metals are transported to the environment by air, water or deposits, thereby increasing the metal concentrations in the environment. For example, the heavy metals concentrations in river waters have been shown to increase several thousand folds by effluents from mining wastes (Agarwal 2009). Therefore, there is probably a

bigger problem with anthropogenically supplied metals, with high levels of bioavailability metals, than with background levels originating from bedrock, with slow weathering (Prasad 2004).

Heavy metal contamination in aquatic ecosystems is a worldwide environmental problem due to its toxicity, persistence and abundance in the environment as well as the subsequent accumulation in aquatic habitats. Generally, the residues of heavy metal in polluted habitats can be accumulated in microorganisms, aquatic fauna and flora, which, in turn, may result in health problems to human through the food chain (Sin et al. 2001). Heavy metals enter rivers from a variety of natural or anthropogenic sources (Gray 2008). Commonly, the smelting and mining activities, industrial effluents contained metal chelates and the indiscriminate use of fertilizer and pesticides including heavy metal in agricultural activities as well as the disposal of untreated and partly treated effluents held toxic metals are the major anthropogenic sources of heavy metal contamination (Martin 2000; Reza & Singh 2010).

The natural processes and anthropogenic impacts generally degrade surface water, and impair their use for drinking, agricultural, industrial and recreational purposes (Krishna et al. 2009). In addition, heavy metals contamination resulting from anthropogenic activities as frequent waste products and their emission often cause the surrounding environment pollution (Eva & Lehikoinen 2000). Under certain environmental circumstances, heavy metals can result in ecological damage and may accumulate to toxic magnitudes (Sekabira et al. 2010). Furthermore, heavy metals also have impacts on various aspects of water use, including water permeability, osmoregulation and oxygen consumption in the aquatic systems by organisms (Ahern & Morris 1998). Evaluation of dissolved heavy metal concentration in river water is necessary to determine the contamination level in the rivers. In aquatic ecosystems, dissolved heavy metals can be immobilized within the river water and can involve complex formation and co-precipitation as hydroxides and oxides of iron and manganese or can occur in particulate matter (Mwiganga & Kansime 2005).

Sungai Semenyih is the main recipient of runoff and effluents for organic substances of domestic effluents and industrial (Al-Badaii & Shuhaimi-Othman 2014; Al-Badaii et al. 2012). Sungai Semenyih is increasingly being contaminated with direct discharge of untreated sewage, raw industrial effluent as well as wastewater from industrial, commercial, and domestic establishments, particularly from Semenyih and Bangi towns (Al-Badaii & Shuhaimi-Othman 2014). Previous studies indicated that the heavy metals concentrations were low in water of Sungai Semenyih (Gasim et al. 2002). In addition, it was reported that the heavy metals concentrations in water of the river is influenced by mining and agricultural and industrial activities as well as deforestation (Al-Badaii et al. 2013). The purpose of this study was to determine the concentrations of dissolved heavy metals in Sungai Semenyih and to use the environmetric methods to evaluate the influences of different pollution sources on heavy metals concentrations.

MATERIALS AND METHODS

STUDY AREA AND SAMPLING SITES

Sungai Semenyih is 266.60 km² in its area and lies between longitude 101° 48'32 .9 "E to 101° 52'30 .5 "E and of latitude 02° 54'14 .9 "N to 03 ° 03'23 .1 "N (Figure 1). The river consists of 25 water catchment valleys and 36 sub-basins with area ranging from 1.37 to 35.57 km² and flows southwards towards the regions of Sepang and Hulu Langat. The climate of study area is characterized by high humidity, high average and homogeneous annual temperatures as well as annual rainfall average about 3000 mm. As a domestic water source, the river supplies 15% of water demand of Kuala Lumpur, Selangor and Putrajaya after the treatment (DOE 2010; Shuhaimi-Othman & Gasim 2005). It is, however, affected by many anthropogenic

activities such as deforestation, settlements and industry, agricultural activities, rubber and oil palm plantations, mining and livestock husbandry farms (Gasim 2003).

Eight sampling stations were selected in this study along the river (Figure 1). Stations 1 and 2 were located in the upstream area. Station 3 was affected by deforestation and mining activity. Station 4 was situated in the area influenced by human activities especially the residential areas. Stations 5 and 6 were in the urban area affected by urban areas and agricultural and industrial activities as well as livestock husbandry farms. Station 7 was located near the construction site of Highway Bridge. The last station was located in Jenderam Hilir after Bangi town and impacted by accumulated pollutants from previous stations, erosion and water treatment plant.

SAMPLE COLLECTION AND PREPARATION

The water samples were collected from the sampling stations in March, July and November of 2012 as well as February, April, July, September and November of 2013. The selection of sampling periods was depending on the different seasons in the study area. The Malaysian Meteorological Department (MMD) (2012) reported that the maximum rainfall in the study area tends to fall in the inter-monsoon months when the equatorial trough lies over Malaysia. The primary maximum takes place during October and November, while the secondary maximum in April and May. In addition, the primary minimum is during June and July and the secondary minimum occurs during January and February. All samples were collected in triplicate using acid-washed polyethylene bottles. The water samples were stored in an iced box at 4°C and immediately transported to the laboratory (APHA 1999). The water samples were filtered in the lab using Whatman filter paper (45 µm) and were acidified to pH2 by Aristar[®] nitric acid (70%) for the analysis of dissolved heavy metals (Fe, Mn, Cd, Cr, Ni, Zn, Cu, Pb and Hg) by Perkin Elmer Sciex ICP-Mass spectrometer Elan 9000. The reason to use ICP-Ms is because it can analyze multiple elements simultaneously and has less chemical interference because of the high plasma temperature as well as less matrix interference because of its mode of sample introduction. In addition, the linearity for ICP-Ms ranges from 4 to 6 orders of magnitude compared to the other instruments that ranges from 2 to 3 orders of magnitude. The ICP-Ms also has variety of emission lines to choose from to increase the sensitivity and to reduce interference from other elements (Gayla 2009; Voica et al. 2012).

QUALITY CONTROL

The quality of analytical data was guaranteed through the execution of laboratory quality control methods, including calibration, analysis of replicates and analysis of blanks. The analysis of duplicate sample analysis was used to determine the consistency in the method. Duplicates (split samples) of the same samples were analyzed identically.

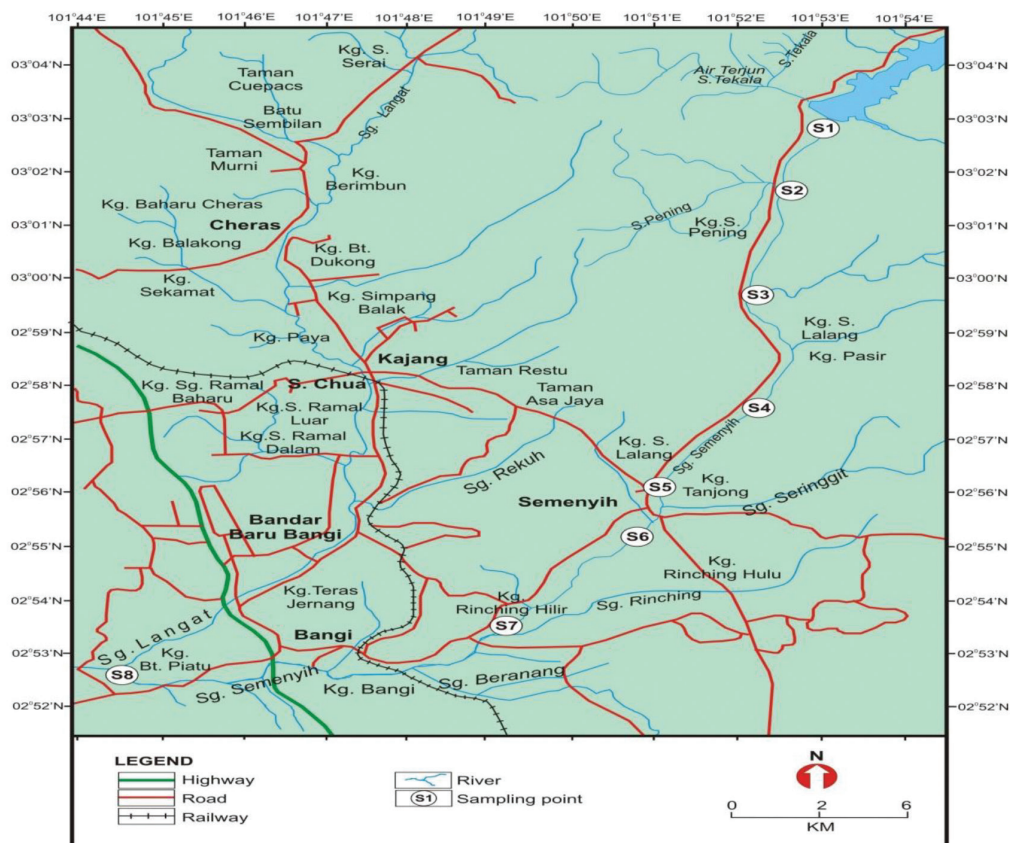


FIGURE 1. Study area and sampling stations along Sungai Semenyih

Then, the difference between the two results of duplicates was calculated (Mitchell 2006). In addition, the analysis of blanks was executed as field blanks and trip blanks. Field blanks were exposed to the sampling environment at the sampling site and handled as the samples to provide information on pollution from all phases of sampling and analysis. The field blanks were prepared immediately after collecting samples at the sampling site (Mitchell 2006). The trip blanks also were performed using bottles filled with blank water (with preservatives) at the lab and transported to and from the field without being opened. The trip blanks were used to check for pollution from the bottles, caps or preservatives. They are particularly valuable for volatile compounds, but also can be used for any type of variable. Trip blanks can become polluted when stored for long time and therefore should be used within one week (Mitchell 2006).

ENVIRONMETRIC METHODS

Environmetric methods were executed using the SPSS version 20, including cluster analysis (CA) and principal component analysis (PCA).

CLUSTER ANALYSIS (CA)

Cluster analysis is an unsupervised pattern recognition method that divides a large group of cases into smaller

clusters of relatively similar cases that are dissimilar to other groups. Hierarchical cluster analysis is the most common approach which starts with each case in a separate cluster and links the clusters together step by step until only one cluster remains (Singh et al. 2004). In cluster analysis, the objects were clustered by linking inter-sample similarities and the outcome showed the overall similarity of variables in the data set (Varol & Şen 2009). In this study, hierarchical cluster analysis (HCA) was executed on the normalized data using Ward's method and squared Euclidean distances as a measure of similarity (Krishna et al. 2009).

PRINCIPAL COMPONENT ANALYSIS (PCA)

Principal component analysis (PCA) can identifies variation in water quality, including heavy metals and determines the effective factors affected on it. Additionally, it extracts the most important variable in polluted stations (Wenning & Erickson 1994). Correlation of original variables and factors generally is given by individual transformed observations and loadings which are called scores (Singh et al. 2004; Zhang et al. 2009). Mathematically, PCA normally involves three main steps: The standardization of measurements to ensure that they have equal weights in the analysis by autoscaling the data to produce new variables, the calculation of the covariance matrix by identifying the eigenvalues and their corresponding eigenvectors and the

elimination of components that account only for a small proportion of the variation in datasets (Vialle et al. 2011).

RESULTS AND DISCUSSION DISSOLVED HEAVY METALS ANALYSIS

IRON (Fe)

The iron concentrations ranged from minimum 315.94 µg/L at sampling station 3 to maximum 636.80 µg/L at sampling station 6 (Table 1). Stations 5, 6 and 7 were affected by the urban runoff and wastewater from Semenyih and Bangi towns that contributed to the high iron concentration. In general, the high iron content is mostly affected by geological formations, particularly from rock-forming silicate minerals such as hornblende, muscovite and biotite (Gray 2008). The content of iron may be associated with iron in rocks, clay soils and argillaceous limestone as well as industrial wastes and mine drainage. Statistical analysis (ANOVA) showed significant differences of iron between sampling stations (ANOVA, $p < 0.05$). In addition, the iron concentrations were lower compared to the results reported by Gasim (2003) in the study of the same basin, which ranged from 591 to 901 µg/L where the high concentrations of iron in Sungai Semenyih were associated with limited sampling station impacted by the rocks and soil weathering, urban storm runoff, discharges of industrial establishments and the runoff from agricultural areas. Compared with various water quality standards (Table 2), the overall mean value of iron in Sungai Semenyih was low compared to the national water quality standard (NWQS). Moreover, it was found to be lower than that of the United States Environmental Agency (CCC), whereas it was higher than the Canadian Standard (CCME) for protecting the aquatic life.

ZINC (Zn)

The zinc values of water samples ranged between minimum 19.58 µg/L at sampling station 4 and maximum 31.90 µg/L at sampling station 5 (Table 1). Adequate zinc is essential to neutralize the toxic influences of cadmium (Varol & Şen 2009). Principally, zinc content has been shown as an example of the evolution of toxic metals related to mining pollution (Olías et al. 2006). In Sungai Semenyih, zinc content is allegedly attributed to industrial and municipal activities (Al-Badaii & Shuhaimi-Othman 2014; Gasim 2003). The statistical analysis using ANOVA showed that there were no significant differences of zinc concentrations between sampling stations (ANOVA, $p > 0.05$). In addition, the zinc concentrations were lower compared to the results reported by Shuhaimi-Othman and Gasim (2005) on the same basin, which were ranged from 14.7 to 70.1 µg/L due to their study was limited by one sampling season, while this study included eight sampling seasons, therefore, they reported that the rock weathering, surface runoff from the surrounding area as well as municipal effluents were the main sources of zinc. The results were also lower than

the results reported by Juahir et al. (2011) on the Sungai Langat basin (range 40-60 µg/L). They reported that the use of metallic roofs coated with zinc and the large number of buildings and houses in the area as the main sources of zinc. This was because when these roofs were in contact with acid rainwater and smog they can readily mobilize zinc into the waterways. As shown in Table 2, the total mean value of zinc in Sungai Semenyih was within the normal level compared to the Canadian Standard (CCME) to protect aquatic life, the United States Environmental Agency (CCC and CMC) and Malaysian standard (NWQS). Thus, in terms of zinc, Sungai Semenyih water is suitable for domestic water supply, agriculture and industrial purposes.

CADMIUM (Cd)

Cadmium is most commonly found associated with zinc in the carbonate and sulfide ores as well as cadmium is widely used in the industrial sector from electroplating to accessory in electronic products (Moore & Ramamoorthy 1984). Cadmium is one of the very dangerous poisons due to cadmium and its compounds are known to result in cancer as well as the negative cadmium effects on the body are numerous and can influence nearly all systems in the body, including gastrointestinal, renal, neurological, cardiovascular, reproductive and respiratory systems (Pitter 1993). The cadmium values were ranged from minimum 0.23 µg/L at sampling station 3 to maximum 0.65 µg/L at sampling station 1 (Table 1). The high content of Cd usually was associated with high fluxes from industrial and urban wastes including the immense urban runoff. In the urban areas, the cadmium content in surface water is affected by using rechargeable batteries in the region (Gray 2008). The statistical analysis using ANOVA showed that there were no significant differences of cadmium concentrations between sampling stations. Moreover, the cadmium concentrations were partly consistent compared to the results reported by Shuhaimi-Othman and Gasim (2005) on the same basin, which were ranged from 0.06 to 0.98 µg/L. Additionally, the result of the overall mean value of cadmium (0.39 µg/L) in Sungai Semenyih was high if compared to the Canadian Standard (CCME) for protecting the aquatic life and the United States Environmental Agency (CCC), whereas it was in the natural level according to Malaysian standard (NWQS), the United States Environmental Agency (CMC) and the World Health Organization (WHO) (Table 2).

MANGANESE (Mn)

In this study, the manganese values of the water samples analyzed were fluctuated from 23.30 to 56.45 µg/L, the maximum value was recorded at the station 8 and the minimum value was obtained at station 3 (Table 1). The manganese content in the water resources is associated with lithologic and anthropogenic sources such as weathering of bedrock, surface run-off, sewage sludge, mining activities as well as manganese can be released to water discharged from industrial facilities or as leachate

from landfills and soil (Prasad 2004). Statistical analysis (ANOVA) found significant differences between sampling stations (ANOVA, $p < 0.05$). In addition, the manganese values were higher than those reported by Shuhaimi-Othman et al. (2009) in the study of Bebar Peat Swampy Forest River, which ranged from 4.2 to 15.3 $\mu\text{g/L}$. The results were also much higher than those obtained by Rosli et al. (2012) in the study of the Sungai Salak basin, which ranged from 0.9 to 9.4 $\mu\text{g/L}$. They reported that the low concentrations of manganese were related to the low industrial activities within their studies areas, whereas in this study the discharge of industrial activities at midstream and downstream stations resulted in high values of manganese, which, in turn affected on the total mean values of the manganese of the river. Finally, the overall mean value of manganese in Sungai Semenyih was in the normal range determined by the Malaysian standard (NWQS) and the World Health Organization (WHO) (Table 2). Therefore, Sungai Semenyih is noted to be suitable for the supply of water for domestic, industrial and agricultural purposes as well as to support the aquatic life.

CHROMIUM (Cr)

The chromium values were ranged from minimum 1.62 $\mu\text{g/L}$ at sampling stations 4 and 7 to maximum 4.80 $\mu\text{g/L}$ at sampling station 8 (Table 1). Moreover, the overall mean concentration of chromium in Sungai Semenyih was 2.37 $\mu\text{g/L}$ (Table 2). Generally, the chromium content is influenced by anthropogenic sources such as industrial effluents derived from the production of corrosion inhibitors and pigments (Gray 2008), which then becomes a pollutant of aquatic ecosystems and thus harmful to aquatic organisms (Shuhaimi-Othman & Gasim 2005). According to the statistical analysis (ANOVA), significant differences of chromium values were found between sampling stations (ANOVA $p < 0.05$). Furthermore, the results were consistent with the results obtained by Lim et al. (2012) on Sungai Langat basin, which ranged from 0.32 to 4.67 $\mu\text{g/L}$, whereas the results were much lower than those reported by Ling et al. (2012) in the study of tropical river (Sungai Serin, Sarawak), which were ranged from 28 to 85 $\mu\text{g/L}$. In comparison with the various water quality standards (Table 2), the total mean value of chromium in Sungai Semenyih was within the normal range compared to the Malaysian standard (NWQS), Canadian Standard (CCME), the United States Environmental Protection Agency (CCC and CMC) and the World Health Organization (WHO). Hence, all the chromium concentrations were found far below the limit and not harmful to use water for any purposes.

NICKEL (Ni)

Nickel is ferromagnetic metal that maintains a high luster and is resistant to corrosion. According to Sunderman and Oskarsson (1991), nickel exists as water-soluble nickel compounds (nitrate and sulfate) and water-insoluble nickel compounds (nickel oxides and nickel hydroxide). In this

study, the nickel values in the sampling stations ranged from minimum 0.53 $\mu\text{g/L}$ at station 3 to maximum 1.06 $\mu\text{g/L}$ at station 8 with a total mean value of 0.79 $\mu\text{g/L}$ (Tables 1 & 2). Commonly, the presence of nickel in the water body is ascribed to anthropogenic activities such as mining, industries and urban wastes (Galvin 1996). The statistical analysis using ANOVA showed that there were no significant differences of nickel concentrations between sampling stations (ANOVA $p > 0.05$). In addition, the nickel content was lower compared to the results reported by Shuhaimi-Othman and Gasim (2005) on the same basin, which were ranged from 1.01 to 7.10 $\mu\text{g/L}$. They reported that the high values of nickel were attributed to the weathering of soils and rocks of the surrounding area as well as the increased industrial activities at midstream (Semenyih town) and downstream (Bangi town). The results were also much lower than those reported by Lim et al. (2012) in the study on Sungai Langat basin (range 0.80-24.72 $\mu\text{g/L}$), who reported that the nickel value was influenced by the receiving of significant quantities of anthropogenic discharges from industrial area, urban areas and agricultural fields, which were related to the electrolyte nickel plating activities, livestock manures, inorganic fertilizer and sewage sludge. The total mean value of nickel in Sungai Semenyih was within the normal range if compared to the Malaysian standard (NWQS), Canadian Standard (CCME) for protecting the aquatic life, the United States Environmental Protection Agency (CCC and CMC) and the World Health Organization (WHO) (Table 2).

LEAD (Pb)

The lead concentrations analyzed in the eight sampling stations ranged from 0.74 to 2.56 $\mu\text{g/L}$, the highest value was recorded at the station 3 and the lowest value was obtained at station 6 (Table 1). Besides, the overall mean value of lead in Sungai Semenyih was 1.62 $\mu\text{g/L}$ (Table 2). Generally, lead deposits in water partitions rapidly between the sediments and an aqueous phase, depending on pH, salt content and the presence of organic chelating agents (WHO 2004). The high content of lead at stations 5, 6, 7 and 8 basically was attributed to the metal group originating from industrial effluents and mining activities. Furthermore, the automobile exhausts are another source of lead, where surface runoff carries the lead deposits in the river (Al-Badaii & Shuhaimi-Othman 2014). Commonly, lead deposited in parking and roads can be carried by surface runoff that flows into the river. In addition, lead is mainly found in water from automobile exhausts through direct atmospheric transport and rainfall. Moreover, statistically significant differences were found between sampling stations for lead concentrations (ANOVA, $p < 0.05$). Besides, the results were higher than those reported by Shuhaimi-Othman et al. (2009) in the study of Bebar Peat Swampy Forest River, which ranged from 0.02-0.066 $\mu\text{g/L}$. The results were also much lower than those obtained by Ling et al. (2012) in the study of tropical River (Sungai Serin, Sarawak), which were ranged from 14 to 61 $\mu\text{g/L}$.

Finally, the overall mean value of lead in Sungai Semenyih was in the normal limits compared to the Canadian Standard (CCME) to protect aquatic life, the United States Environmental Agency (CCC and CMC), the World Health Organization (WHO) and the Malaysian standard (NWQS) (Table 2). Therefore, in terms of lead, Sungai Semenyih is suitable for the support of aquatic life and water domestic supply as well as industrial and agricultural uses.

COPPER (Cu)

Copper is one of the several heavy metals that are essential to life despite being inherently toxic as non-essential metals exemplified by lead and mercury (Scheinberg 1991). In this study, the copper values ranged from minimum 1.89 µg/L at sampling station 1 to maximum 15.69 µg/L at sampling station 8 with total mean value of 7.72 µg/L (Table 1). The anthropogenic sources related to the discharge of domestic sewage and industrial effluents generally result in pollution with copper in receiving water. The high content of copper at stations 2, 4 and 8 may come from leaching processes of country rocks as well as the surface run off from a palm plantation area located before station 2 which may be use as a fungicide and pesticide. According to USEPA (2012), copper may be released into the water ecosystem through agricultural activities that use copper as a pesticide, fungicide and an algacide. At downstream area, the high content of copper was found to be related to farming and agricultural and industrial activities and surface runoff for deforested areas. According to USEPA (2012), copper may enter the

water environment through industrial activities such as leather industry and leather products, fabricated metal products, electrical equipment and automobile brake pads as well as the natural processes such as decaying vegetation, surface runoff, windblown dusts and forest fires which distributed high content of copper. Statistical analysis showed that there were no significant differences between sampling stations for copper content (ANOVA $p>0.05$). In addition, the copper values were lower than the results obtained by Shuhaimi-Othman and Gasim (2005) in the study on the same basin, which ranged from 5.96 to 21.2 µg/L. They stated that the high value of copper was associated with farming and agricultural activities, where it was used as a pesticide and support for growth in the livestock farms as well as their study was limited to March and affected by the weathering of rocks at upstream stations and urban runoff and industrial wastewater at the downstream stations. Whereas, in this study, the low value of copper was ascribed to the dilution of river water during the rainy season due to this study including eight seasons. In comparison with the various water quality standards (Table 2), the total mean value of copper in Sungai Semenyih was higher compared to the Canadian Standard (CCME) to protect the aquatic life, while it was in the normal level compared to the United States Environmental Agency (CMC and CCC), the Malaysian standard (NWQS) and the World Health Organization (WHO). In conclusion, Sungai Semenyih system was suitable for the support of aquatic life and water supply for domestic, industrial and agricultural uses with regards to copper content.

TABLE 1. Mean values of dissolved heavy metals in March, July, November of 2012 and February, April, July, September, November of 2013 of Sungai Semenyih

Stations, n= 8	µ	Zn	Cd	Mn	Cr	Ni	Pb	Cu	Hg	
1	Mean	336.12	23.63	0.65	30.15	2.89	0.88	1.26	1.89	0.12
	SD	123.82	15.34	0.55	27.60	1.39	1.06	0.94	0.90	0.18
2	Mean	345.27	21.79	0.26	25.10	1.75	0.63	0.87	9.19	0.14
	SD	66.63	29.79	0.32	13.57	1.49	0.71	0.38	14.89	0.13
3	Mean	315.94	19.49	0.23	23.30	2.35	0.53	0.74	5.88	0.18
	SD	77.32	28.23	0.16	12.75	2.16	0.58	0.27	8.51	0.23
4	Mean	367.39	19.58	0.41	25.15	1.62	0.63	1.25	13.69	0.24
	SD	79.61	19.69	0.66	10.44	1.53	0.52	0.96	22.23	0.19
5	Mean	540.26	31.90	0.59	28.02	1.83	0.89	1.93	8.30	0.65
	SD	232.06	32.96	0.53	10.26	1.00	0.82	1.21	14.24	0.94
6	Mean	636.80	23.83	0.28	40.22	2.13	0.95	2.56	4.52	0.25
	SD	242.52	12.50	0.23	7.95	1.30	0.80	1.84	5.30	0.12
7	Mean	534.42	29.39	0.25	44.01	1.62	0.75	2.12	2.61	0.30
	SD	342.18	31.93	0.23	19.17	1.23	0.54	1.16	2.03	0.26
8	Mean	456.49	28.64	0.46	56.45	4.80	1.06	2.25	15.69	0.30
	SD	286.86	28.61	0.33	27.27	2.74	0.77	1.83	23.43	0.20

MERCURY (Hg)

Mercury is widely used in disinfectants, fungicides (in wood, timber preservations and paints) antiseptic, dental amalgam, herbicides and in scientific instruments and electrical equipments (Van Burg & Greenwood 1991). The mercury values in the sampling stations ranged from 0.12 to 0.65 µg/L (Table 1). The highest value was recorded at station 5, whereas the lowest was recorded at station 1 (Table 1). In addition, the total mean value of mercury in Sungai Semenyih was 0.27 µg/L (Table 2). The mercury content in Sungai Semenyih was associated with rocks, sediments and soils which naturally contain small but varying amounts of mercury. In addition, the use of mercury in the timber preservation in timber mills (Fortin et al. 2001) may be one of the sources of mercury in the river due to there are some timber mills found in the study area particularly at station 5. The statistical analysis (ANOVA) found that there were no significant differences of mean mercury values between sampling stations (ANOVA $p > 0.05$). In comparison with the various water quality standards (Table 2), the overall mean value of mercury in Sungai Semenyih was within the normal range determined by the NWQS for Malaysian

ivers. Additionally, the overall mean value of mercury in Sungai Semenyih was in the natural limits if compared to the United States Environmental Agency (CMC and CCC) and the World Health Organization (WHO). Therefore, in terms of mercury, Sungai Semenyih water is suitable for domestic, agricultural and industrial purposes.

CLUSTER ANALYSIS

Cluster Analysis (CA) using Ward's method with squared Euclidean distances was used to determine the similarities between the sampling stations (Sundaray et al. 2011; Varol & Şen 2009; Zhang et al. 2009). It generated two statistically significant clusters of the eight sampling stations at $(D_{link}/D_{max}) * 25 < 5$ as represented in a dendrogram (Figure 2). Cluster 1 consisted of stations 1, 2, 3 and 4, whereas cluster 2 comprised of stations 5, 6, 7 and 8. In cluster 1, station 1 was situated in the area covered by forest in the upstream. Generally, the upstream area of rivers is covered with intensely forest and influenced only by surface runoff (Al-Badaii & Shuhaimi-Othman 2014; Al-Badaii et al. 2013; Bu et al. 2010). At this station, anthropogenic activities were limited except for some recreational activities at waterfall

TABLE 2. Criteria of heavy metals concentrations in freshwater ecosystem

Metals	Present study µg/L	CCME µg/L	USEPA-(CMC) µg/L	USEPA-(CCC) µg/L	WHO µg/L	NWQS, class II (µg/L)
Fe	441.59	300	-	1000	-	1000
Zn	24.78	30	120	120	-	5000
Cd	0.39	0.017	2.0	0.25	3	10
Mn	34.05	-	-	-	400	100
Cr	2.37	8.9	570	87	50	50
Ni	0.79	52-150	470	52	70	50
Pb	1.62	1-7	65	2.5	10	50
Cu	7.72	2-4	13	9.0	2000	20
Hg	0.27	-	1.4	0.77	1	1

Source: [34, 41, 46, 47]

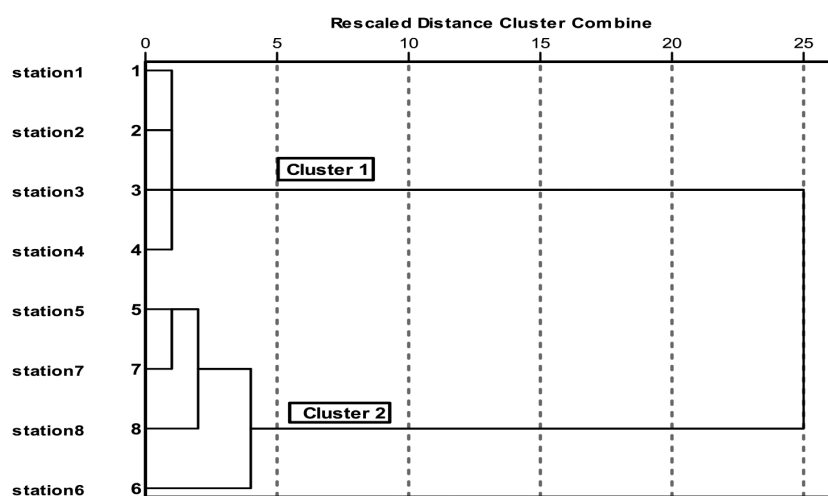


FIGURE 2. Dendrogram showing clustering of sampling stations in Sungai Semenyih

in upstream. Besides, station 2 was located near to station 1 and influenced by deforested areas and palm plantation. Furthermore, stations 3 and 4 receive pollution from mining, domestic effluents of unsewered from residential areas and agricultural activities. In cluster 2, the sampling stations had similar characteristic which was impacted by mainly parallel pollution sources in consequence of the land use changed of forest to agriculture, unplanned settlements and livestock farms (Al-Badaii & Shuhaimi-Othman 2014). Furthermore, the stations of cluster 2 were located in the middle and downstream of the river and influenced by urban runoff and anthropogenic activities. Station 5 was impacted by the industrial activities, surface runoff and wastewater from Semenyih town (Al-Badaii et al. 2013, 2012). Station 6 appeared to be principally influenced by the livestock farms and leaches from small dumps situated on the banks of river course. Besides, stations 7 and 8 were situated in the downstream and influenced by deforestation, rubber and oil palm plantation, agricultural and industrial activities, storm water runoff, discharge of vehicles washing, erosion and workshops as well as station 8 was affected by the pollution coming from industrial area of Nilai loaded by Sungai Pajam and Sungai Beranang. Overall, anthropogenic activities in this area comprised of industries and settlements, which covered Bangi, Rinching, Beranang and Broga, caused deterioration of river water (Shuhaimi-Othman & Gasim 2005). These results exhibited that Sungai Semenyih, particularly in the midstream and downstream stations was impacted by untreated domestic effluents, livestock husbandry, agricultural and urban runoff, and industrial activities besides discharge of domestic wastewater from Bangi and Semenyih towns that includes more than half the population around Semenyih basin (DOE 2010). In addition, the results point to, that cluster analysis is a positive environmetric technique to categorize and evaluate the surface water in Sungai Semenyih basin. At the same time, it is a significant technique to build an optimal strategy to reduce sampling stations and experimental analysis

cost. Eventually, the cluster analysis classify that Sungai Semenyih based on the similarity of sampling stations into two areas, low pollution area (stations 1, 2, 3 and 4) and high pollution area (stations 5, 6, 7 and 8).

PRINCIPAL COMPONENT ANALYSIS/ FACTOR ANALYSIS

Principal component analysis/factor analysis was separately carried out on the normalized data sets for the two different areas (low pollution area and high pollution area) as described by the cluster analysis technique to recognize the factors affecting on each area (Krishna et al. 2009; Razmkhah et al. 2010; Sekabira et al. 2010; Varol & Şen 2009). Factor analysis of the two data sets yield two factors for low pollution area (Table 3) and three factors for the high pollution area (Table 4) with Eigenvalues >1 (Figures 3 and 4), representing 92.544% and 100% of the total variance in each heavy metals data sets. Generally, the Eigenvalue gives a measure of the factor significance for the factor. Eigenvalues of 1.0 or greater are significant, whereas Eigenvalues which were less than 1 are insignificant (Varol & Şen 2009). Additionally, the factor loadings were categorized based on the loading values as strong (>0.75) moderate (0.75–0.50) and weak (0.50-0.30) (Zhang et al. 2009).

For the data set of low pollution area, factor 1 elucidated 49.946% of the total variance with strong positive loadings on Cd, Mn, Ni, Pb and moderate loading on Zn. The positive loading of Cd is affected by the granite body of the upstream areas that contain cadmium in high levels because according to Gasim (2003), the upstream area of Sungai Semenyih is covered by granitic body that contain high content of Cd, therefore, the weathering of rocks and soils in this area has caused the presence of Cd in high concentrations in the upstream of river. In addition, the presence of Mn could be ascribed to the surface runoff from surrounding area and rocks weathering due to according to Van Ha et al. (2011) and Ziemacki et al. (1989), the Mn is very abundant metals that is widely distributed in the soils in relation to oxides, silicates and

TABLE 3. Factor loading of the nine dissolved heavy metals on Varimax rotation at low pollution area in Sungai Semenyih

Heavy metals	Factor	
	1	2
Fe	0.463	0.855
Zn	0.712	-0.577
Cd	0.956	-0.185
Mn	0.926	-0.377
Cr	0.359	-0.853
Ni	0.930	-0.365
Pb	0.917	0.341
Cu	-0.240	0.960
Hg	-0.265	0.858
Variance %	49.496	43.048
Cumulative %	49.496	92.544

TABLE 4. Factor loading of the nine dissolved heavy metals on Varimax rotation at high pollution area in Sungai Semenyih

Heavy metals	Factor		
	1	2	3
Fe	-0.592	-0.780	0.201
Zn	-0.021	0.919	0.394
Cd	0.470	0.390	0.792
Mn	0.586	0.035	-0.809
Cr	0.955	0.072	-0.288
Ni	0.936	-0.340	0.085
Pb	-0.149	0.895	0.423
Cu	0.968	0.236	0.088
Hg	-0.074	0.467	0.881
Variance %	40.479	31.130	28.121
Cumulative %	40.479	71.879	100.000

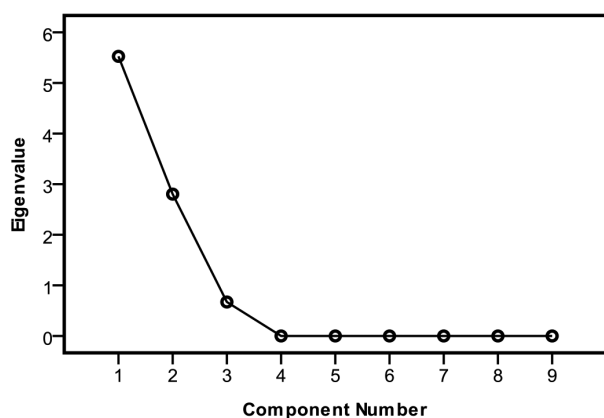


FIGURE 3. Scree plots of the factor model of the low pollution area

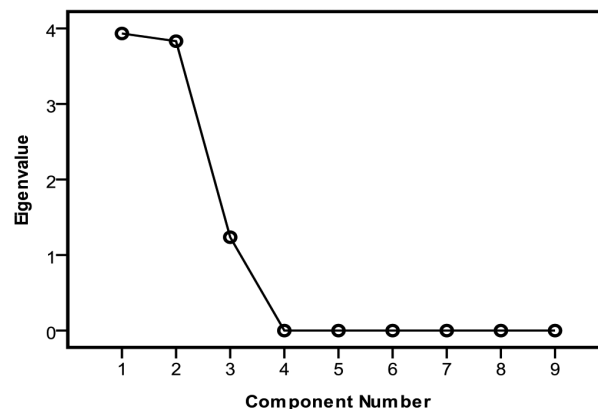


FIGURE 4. Scree plots of the factor model of the high pollution area

carbonates, which reach the water resources during the storm water events. The positive loading of Ni could be affected by the natural sources related to the weathering of rocks, soil erosion and the vegetation because according to Al-Badaii and Shuhaimi-Othman (2014), the Ni level in the upstream area of Sungai Semenyih was associated with metamorphic rocks contained high content of Ni. According to Cempel and Nikel (2006), the Ni is widely distributed in the environment, being released from both anthropogenic activity and natural sources derived from the weathering of rocks and soils, forest fires and decaying of vegetation. Furthermore, the presence of Pb was influenced by the surface runoff from the deforested area and sand mining due to according to Gasim (2003) the granitic soils is one of the sources for Pb in Sungai Semenyih. The moderate positive loading of Zn content could be attributed to the geological characteristics of the upstream area due to according to Al-Badaii and Shuhaimi-Othman (2014), the Zn content in Sungai Semenyih was related to the soils from the Semenyih Granite series that contain high content of Zn, metamorphic soils and the quartzite rock. Zakir et al. (2006) emphasized that the largest portion

of heavy metals such as Zn in natural water resources is usually tied up primarily in two forms as adsorbed on the surfaces of particulate material like organic debris or clay and weathered solids of precipitates. Factor 2 explained 43.048% of the total variance, has strong positive loading on Fe, Cu and Hg, whereas it has strong negative loading on Cr and moderate negative loading on Zn. the strong positive loading of Fe is associated with iron in rocks, clay soils and argillaceous limestone as well as industrial wastes and mine drainage (Agarwal 2009). Moreover, the strong positive loading of Cu is ascribed to the weathering of rocks and soil as well as leaching from ore/rocks and from oil waste. According to Kumar et al. (2013), the Cu was released into water due to the natural weathering of soil, mining and industrial discharges as well as the presence of Hg content in Sungai Semenyih was associated with rocks, sediments, water and soils which naturally contain small but varying amounts of Hg (Gasim 2003). Therefore, in low pollution area, factors 1 and 2 stands for lithologic sources of pollution.

For the data set of high pollution area, factor 1 explained 40.749% of the total variance strong positive

loading on Cr, Ni, Cu and moderate positive loadings on Mn. Generally, the presence of Cr is affected by anthropogenic sources such as industrial effluents derived from the production of corrosion inhibitors and pigments which then becomes pollutants of aquatic ecosystems and thus harmful to aquatic organisms (Agarwal 2009). The presence of Cr in Sungai Semenyih was impacted by the urban area at Semenyih town where the runoff contains chromium from paints on buildings and detergents in wastewater as well as the industrial effluents from Bangi town and Nilai industrial area. Rahman et al. (2014) reported that the main sources of chromium were industrial wastes related to tannery wastes, chromium pigment, leather manufacturing wastes and municipal sewage sludge. Additionally, the strong positive loading of Ni is related to industrial wastewater from Semenyih town as well as the agricultural runoff in the downstream area. Generally, nickel compounds have many industrial and commercial uses such as the stainless steel production and nickel alloys with high corrosion and temperature resistance (Cavani 2005). In addition, the presence of Ni is ascribed to the domestic wastewater and industrial discharges from Bangi town and Nilai industrial area as well as the agricultural activities where Ni is applied in agricultural activities due to phosphate fertilizers which include traces of Ni (Cempel & Nikel 2006). Besides, the anthropogenic sources related to the discharge of domestic sewage, agricultural activities, animal farms and industrial effluents generally result in pollution with Cu in receiving water (Agarwal 2009). The Cu may enter the water environment through industrial activities such as leather industry and leather products, fabricated metal products, electrical equipment and automobile brake pads (USEPA 2005). Generally, Cu is used in mineral supplements to correct Cu deficiencies in farm animals and to stimulate growth and fattening of pigs and poultry (Al-Badaii & Shuhaimi-Othman 2014). Therefore, this factor corresponds to the contribution of the industrial and agricultural activities and electroplating wastewater in the water pollution. Factor 2 indicated 31.130% of the total variance with strong positive loading on Zn, Pb and a strong negative loading on Fe. This factor indicated pollution from domestic wastewater particularly at Semenyih and Bangi towns. Generally, the strong loading of Zn is attributed to add the Zn to the environment from the industrial activities such as mining, steel processing and waste combustion (Edwards 2010). In addition, the strong loading of Pb is ascribed to the discharge of Pb into surface water through solders, paints, pipes, gasoline and building material. Factor 3 explained 28.121% of the total variance and has strong positive loadings on Cd and Hg, whereas it has moderate negative loading on Mn. The strong positive loading of Cd was associated with the urban area at Semenyih and Bangi towns which were affected by the continuous development. In the urban areas, the Cd content in surface water is affected by using rechargeable batteries as well as the using of Cd in pigments, plating and coatings and as stabilizers for plastics (Agarwal 2009).

In addition, the strong positive loading of Hg could be impacted by the industrial activities in urban area due to (Kowalski et al. 2007) the mercury sources in urban areas were related to combustion facilities, including coal-fired power plants, hospital incineration and municipal solid waste incineration as well as sewage sludge and hazardous waste. Finally, in high pollution area, the industrial discharge, urban runoff, agricultural activities and animal farms which consider either point or non-point source pollution is the main sources of pollution with metals.

In this study, factor analysis/principal component analysis of low pollution area did not result in much data reduction, as we need 8 variables that represents 88.889% of the 9 variables to elucidate 92.544% of the data variance as well as factor analysis of high pollution area did not results in much data reduction, as we need 8 variables that represents 88.889% of the 9 variables to explain 100% of the data variance. Nevertheless, factor analysis served as a means to recognize those variables, which have great contribution to variation in the dissolved metals in the river. Similar techniques based on factor analysis were used to assess the heavy metals variations in the rivers (Krishna et al. 2009; Razmkhah et al. 2010; Zhang et al. 2009).

CONCLUSION

In this case study, the measurement of dissolved heavy metals in water samples of Sungai Semenyih indicated that metals were in the normal range determined by the NWQS. In addition, Hierarchical cluster analysis using Ward's method with squared Euclidean distances classified 8 sampling stations into two clusters (low pollution area and high pollution area) based on the similarity of sampling stations characteristics. This classification is valuable to build an optimal strategy to reduce sampling stations and experimental analysis cost. The principal component analysis/ factor analysis extracted and identified the factors responsible for dissolved heavy metals variations in the river at two different pollution areas. Nevertheless, PCA/FA of low pollution area did not result in significant data reduction, as we need 8 variables that represents 88.889% of the 9 variables to elucidate 92.544% of the data variance as well as factor analysis of high pollution area did not result in much data reduction, as we need 8 variables that represents 88.889% of the 9 variables to explain 100% of the data variance. However, factor analysis served as a means to recognize those variables, which have great contribution to variation in the dissolved metals in the river. Based on the results obtained from factor analysis, the anthropogenic (agricultural activities) and lithologic (bedrocks weathering) sources were responsible for dissolved metals variations in the low pollution area, whereas in the high pollution area, the anthropogenic sources such as domestic wastewater, agricultural activities, industrial effluents and animal farms were responsible for dissolved metals variations. Therefore, this study showed the usefulness of environmetric methods to evaluate and interpret the complex data sets as well

as it stated its importance for identification of pollution factors and understanding variations in dissolved metals in the river.

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