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Analyses and Pollution Potential of heavy metals at The Jerangau-Jabor Landfill in Kuantan, Malaysia

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ABSTRACT: The impact of Industrialization has always been related to the better economic and social transformation. However, it should be well planned for environmental sustainability. Landfilling is the most used municipal solid waste (MSW) disposal method in Malaysia. Raw and treated leachate collected from Jerangau-Jabor Landfill Site (JJLS), Kuantan, Pahang were analysed for the content of silver, cadmium, chromium, copper, iron, lead, zinc using Flame Atomic Absorption Spectrometry (FAAS) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The metal analyses result were compared with standard limits from the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 (Act 127) set by the Department of Environment, Ministry of Natural Resources and Environment, Malaysia and used to calculate the sub-leachate pollution index of heavy metals (sub-LPIhm) to evaluate the pollution potential of the heavy metals. The sub-LPIhm is one of the sub-index needed to calculate the Leachate Pollution Index (LPI) together with the sub-LPI organic (sub-LPIorg) and sub-LPI inorganic (sub-LPIinorg). LPI is the level of leachate pollution potential of a landfill site. All the heavy metals in the raw leachate were significantly higher than the treated leachate. Some were found to be above the permissible standard limit stipulated in the regulation. However, the sub-LPIhm showed that the level of heavy metal pollution potential of the leachate is low. It is recommended that the treated leachate should undergo continuous treatment to ensure the discharge leachate complied with the standard limit.

Keywords: Heavy metals; Landfill leachate; Sub-leachate pollution index of heavy metal; Pollution potential.

INTRODUCTION

Landfills are the most widely utilized solid waste management option in Malaysia with 94.5% of municipal solid waste (MSW) been disposed at the landfill (Tan et al., 2014). Kuantan, the capital of the state of Pahang is located in the east coast of Malaysia shown in Figure 1. It is well known for its beautiful beaches and at the same time, an industrial hub that houses The Semambu Industrial Estate, The Kuantan Port Industrial Area and The Gebeng Integrated Petrochemical Complex (Figure 1). Kuantan Jerangau-Jabor Landfill Site (JJLS) is a sanitary landfill with the concept of ‘Semi-aerobic Re-circulatory System’ or ‘Fukuoka Method’ located in Kuantan that received all generated MSW. In record, the amount of solid wastes produced in Kuantan is about 500 tons daily, consisting of 300 tons domestic and 200 tons industrial

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and construction wastes (Zahari et al., 2010). The industrial wastes vary from chemical, petrochemical, palm oil, manufacturing and engineering industries. It is expected that this amount of waste would possibly increase with the full operation of the Malaysia-China Kuantan Industrial Park (MCKIP) as indicated in Figure 1, which includes steel and non-ferrous metals industry, clean technology and renewable energy, petrochemical industry, research and development, electrical and electronic energy and machinery and equipment manufacturing.

Fig. 1. Location of Kuantan, Kuantan Port and the Malaysia-China Kuantan Industrial Park (MCKIP) [Source: Malaysiankini.com and Kuantanport.com.my/en_GB]

One of the major problems with landfilling is the generation of landfill leachate. Leachate is the liquid generated from water that infiltrates the stored waste within the landfill. It is well known that landfill leachates are capable to cause serious environmental pollution that includes surface water, groundwater and soil pollution killing plants, animal and humans because they can be highly toxic with large quantities of organic and inorganic matters and high concentration of heavy metals. Heavy metals are generally known carcinogenic and mutagenic for living organisms if exceeded the permissible limit. They are capable to form stable complexes that increase their mobility in leachate-polluted waters (Mukherjee et al., 2015). Quantification of the level of contamination have played an important element in the monitoring the metallic pollution of the river water using heavy metal pollution index (HPI) and contamination index (Cd) (Nasrabadi, 2015), metal pollution in agriculture soil using detailed statistical analysis (Karbassi et al., 2014) and dissolved and particle-bound metal fluxes in streams can be evaluated using linear relationships between bulk metal concentrations in water and total suspended solids in water (Nasrabadi et al., 2018)

Our approach towards environmental pollution has leads us to investigate, compare the content of heavy metals and access the condition of the landfill. The concentration of heavy metals were assess against the standard value limits obtained from Environmental Quality (Control of

Pollution From Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 (Act 127). The condition of a landfill can be expressed from the level of leachate pollution potential of a landfill site, which can be calculated using leachate pollution Index (LPI) formulated using Rand Corporation Delphi Technique (Kumar & Alappat, 2003). The LPI is formulated from calculated sub-leachate pollution indices based on the categorized list of dominant group of pollutants, which are sub-LPI organic (sub-LPIorg), sub-LPI inorganic (sub-LPIinorg) and sub-LPI heavy metals (sub-LPIhm) (Kumar & Alappat, 2005). The LPI value is an increasing scale index of a single number ranging from 5 to 100, wherein a higher value indicates a poor environmental condition. A minimum value of 5 of leachate pollution is considered to ensure that a multiplicative aggregation function could be used, if required, and the minimum value of 5 units of leachate pollution will ensure that the LPI value does not result in zero even if some of the pollutants do not show any pollution. Therefore, the theoretical range of LPI is from 5 to 100 (Kumar & Alappat, 2004).

MATERIAL AND METHODS

All chemicals were of analytical grade and were used without further purification. Leachate samples were collected from leachate treatment plant (LTP) of the Jerangau-Jabor, Kuantan landfill which located at 3^{o} 56’ 53”N, 103^{o} 21’ 3”E, along the al-Muktafi Billah Shah Road, Kuantan road. All samples were collected in the month of May to July 2017 and taken from the depth of 1 meter. Leachate samples collection was done at the leachate collection pond (leachate before treatment shown in Figure 2) and the tertiary feed chamber (leachate after treatment shown in Figure 3) where the treated leachate was accumulated. All the samples were collected in triplicate using amber bottles and immediately acidified with a few drops of concentrated nitric acid. The samples were stored at 4 °C upon waiting for digestion. The description of the leachate samples are raw leachate collected from the inlet pipeline of the landfill (RL), treated leachate collected from the inlet pipeline after treatment (TL1), treated leachate collected from the outlet pipeline of the leachate discharged point (TL2) and treated leachate collected near the submersible pump in the chamber (TL3).

Fig. 2. Raw leachate collected from leachate collection pond

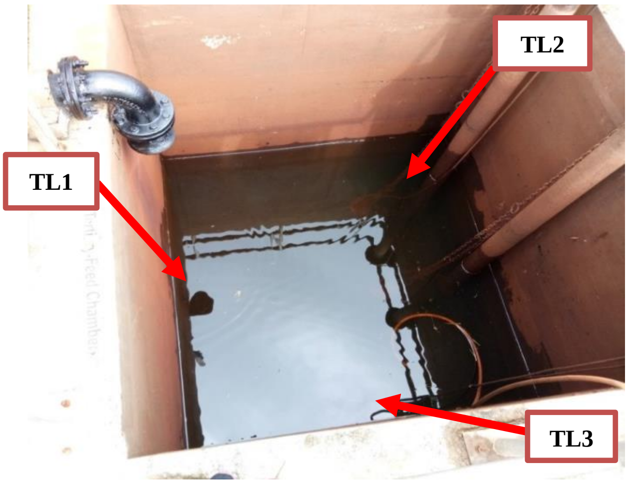

Fig. 3. Treated leachate sample collected from tertiary leachate chamber

The samples, which were kept at 4 °C were thawed and left at room temperature for about 3 hours prior to digestion. All samples were digested using microwave assisted acid digestion according to USEPA Method 3015A (U.S. EPA., 2007). The concentration of chromium and cadmium were analyzed using Flame Atomic Absorption Spectrometer (FAAS) with the correlation coefficient of 0.9995 and 0.9996, respectively. The concentration of silver, copper, iron, lead and zinc were determined using Perkin Elmer Inductively Coupled Plasma-Mass Spectroscopy Series 200 auto sampler. The correlation coefficient of the calibration curve were 0.996942 for silver, 0.996786 for copper, 0.998576 for iron, 0.999820 for lead and 0.997074 for zinc. All analyses were done according to U.S. EPA Method 6020B to ensure method performance and result validation (U.S. EPA., 2014).

The sub-LPI for heavy metal was formulated based on the Leachate Pollution Index (LPI) as in the Eq. 1 based upon five selected heavy metals were chromium, lead, zinc, copper and iron, which possess different weight factors (w_i) of a wide range of significant levels as reported earlier (Kumar & Alappat, 2004). The weight factor indicates the importance of each pollutant variable to the overall leachate pollution. Chromium and lead were of high importance with the highest weight factor of 0.064 and 0.063, respectively. Zinc has a moderate weight factor of 0.056. Copper and iron were of least importance with a low weight factor of 0.050 and 0.045, respectively. After the concentration of the heavy metals were analysed and known, the sub-index values (p_i) were determined based on the sub-index curves for all the variable pollutant as reported by Kumar & Alappat (2003). The weighted sum linear aggregation function was used to sum up all the parameter of the leachate pollutant variables. The various possible aggregation functions were evaluated by Kumar & Alappat (2004) to select the best possible aggregation function for the existing pollutant variables. The sub-LPIhm was calculated as in the Eq. 2.

$$LPI = \frac{\sum_{i=1}^{n} w_i P_i}{\sum wi} \tag{1}$$

Where LPI = leachate Pollution Index

w_i = the weight for the ith pollutant variable,

p_i = the sub index value of the $_i$th leachate pollutant variable,

n = number of leachate pollutant variables for which data is available and $\sum wi < 1$

$$sub - LPI\left(hm\right) = \frac{\sum_{i=1}^{n} w_i P_i}{\sum wi} \tag{2}$$

Where sub-LPI(hm) = the weighted additive of sub-index leachate Pollution Index heavy metal

w_i = the weight for the *i*th pollutant variable,

p_i = the sub index value of the $_i$th leachate pollutant variable,

n = number of leachate pollutant variables for which data is available and $\sum wi < 1$

A thorough literature search was made to collect information on various landfills across Malaysia within a period of 10 years. All the raw data were tabulated for comparison with data from this work and the standard value limits obtained from Environmental Quality (Control of Pollution From Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 (Act 127) in Table 2. The raw data (Table 2) were utilized to calculate the sub-leachate pollution index of heavy metals (sub-LPIhm) based on the same method (as mentioned in section 2.3) and tabulated in Table 3 for comparison of the changes of the various landfills across Malaysia within a period of 10 years.

RESULT AND DISCUSSION

In Malaysia, the quality of the leachate discharged from any sanitary landfill system is bound by Environmental Quality (Control of Pollution From Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 (Act 127) with other standards and by-laws adopted by the local authorities. Therefore, the leachate discharge from sanitary landfill system must be treated to comply with the standard limits. Table 1 showed that the metal with the highest concentration found in the raw leachate was Fe, followed by Cr, Zn, Cd, Cu, Pb and Ag. While, the treated leachate exhibited highest concentration of Fe, followed by Cr, Cd, Zn, Pb, Cu and Ag as recorded in Table 1. The concentration of cadmium, chromium and iron in JJLS were found to exceed the standard permissible limits. The heavy metals concentration pattern and distribution in JJLS were resulted from the 60% domestic waste and 40% of industrial and construction waste deposited at the landfill (Zahari et al., 2010). Variation of the values may be due to the differences in waste composition, moisture content, site hydrology, topography of landfill site, waste compaction, interaction of leachate with the environment, etc.

Table 1. Concentration of heavy metals in raw and treated leachate

Heavy metals	Raw leachate (mg/L)	Point 1	Treated leachate (mg/L)		Average	Percentage of reduction (%)	Acceptable conditions for the discharge of leachate*
			Point 2	Point 3			
Ag	0.0041± 0.0010	0.0023± 0.0004	0.0007± 0.0001	0.0028± 0.0009	0.0024± 0.0017	41.46	0.10
Cd	0.1078± 0.0110	0.0594± 0.0171	0.0444± 0.0069	0.1778± 0.1128	0.0706± 0.0331	34.51	0.01
Cr	0.4956± 0.1305	0.7100± 0.0324	0.7500± 0.0495	0.7944± 0.0083	0.7515± 0.0422	+51.63	0.05# and 0.20
Cu	0.0333± 0.0104	0.0070± 0.0004	0.0075± 0.0013	0.0043± 0.0005	0.0063± 0.0017	81.08	0.20
Fe	6.0606± 0.6439	6.0374± 2.3513	3.3849± 0.0851	3.6774± 0.7145	4.3665± 1.4544	27.95	5
Pb	0.0261± 0.0038	0.0183± 0.0039	0.0105± 0.0094	0.0033± 0.0003	0.0107± 0.0075	59.00	0.10
Zn	0.1882± 0.0618	0.0492±0.0084	0.0035± 0.0014	0.0159± 0.0016	0.0229± 0.0237	87.83	2.0

*Environmental Quality (Control of Pollution From Solid Waste Transfer Station and Landfill) Regulations 2009 (PU(A) 433)
#concentration of hexavalent chromium

Generally, it is found that the heavy metals are highly concentrated near the submersible pump (TL3) as shown in Table 1. This could be explained because the driving force of the pump to pull water into the pump that later pushes out the water to the surface affected the distribution of heavy metals in the leachate. Therefore some metals may accumulate near the pump. The concentration of heavy metals was higher near the inlet pipeline after the treatment pond (TL1) and gradually become lower near the outlet pipeline upon the discharge of leachate (TL2).

It was found that all the treated leachate contained lower concentration of heavy metals than the raw leachate, except for chromium, which its concentration was increased about 51.63% in the treated leachate as shown in Table 1. There are many ways that chromium could be present in landfill because it is widely used in pigments and dyes, electrolytic chromium plating, cement, leather tanning, and wood preservation industries (OSHA, 2006). However, the main contributing factor is the high concentration of chromium was found in deposited soil around stockpile and bauxite mining area in Kuantan, which derived from the crustal mineral, mine waste or residues as well as dust and aerosol emission from the extraction, transportation and deposited of soil particles in the mining area (Syed Ismail et al., 2018).

The combination of biological, chemical and physical treatment applied at the Kuantan landfill's LTP has proved to be efficient and effective to remove other heavy metals in the leachate treatment process. The underlying piping system allows air flow inside and outside the waste through leachate collection pipes by passive ventilation to accelerate aerobic microbial decomposition in the waste. The leachate will undergo decantation, coagulation and flocculation processes to remove the precipitates and sludge before the treated leachate flow into the tertiary feed chamber (Al-Aziz, 2013).

The concentration of zinc displayed a most significant reduction of 87.83% in treated leachate followed by copper and lead with a percentage reduction of 81.08% and 59.00%, respectively. Zinc deposited in landfill attributed to mining activities, burning of coal, scrape metals, dry cell batteries, paint, dyes, ceramics and rubber products (Adeolu et al., 2011; Moturi et al., 2004). The presence of copper proved the disposal of considerable amount of paints, blades, bottle caps, insecticides, pharmaceuticals and cosmetics (Kanmani & Gandhimathi, 2013). Lead is found due to the disposal of lead batteries, lead based paints, chemicals from photograph processing, plastics, and pipe in the site (Moturi et al., 2004).

Silver, cadmium and iron in treated leachate also exhibited a percentage concentration reduction of 41.46%, 34.51% and 27.95%, respectively. Photographic industries produced considerable amount of waste containing silver (Orubite-Okorosaye & Jack, 2012). Waste containing cadmium usually found the batteries, electronic devises, ceramics and glass, pigments and plastics (Tamaddon & Hogland, 1993). The presence of iron attributed to the iron-based material waste from construction materials such as paints, pigment, polishing agents, electrical appliances and steel scrap (Aziz et al., 2004).

A comparison was drawn from the result with previous data from all the landfills around Malaysia within a period range of 10 years in Table 2 showed that the concentration of heavy metals in all the landfills varied with the different quality of leachate due to the various type and composition of waste deposited in that landfill. All the landfill has one or more heavy metals that is highly concentrated and exceeded the standard limits according to the Malaysian Environmental Quality (Control of Pollution from Solid Waste Transfer

Station and Landfill) Regulation 2009. Jeram Landfill consist about 95% domestic waste and 5% industrial waste displayed concentration of cadmium, chromium, iron and zinc beyond the standard limits. The concentration of lead was found to be above the standard limit in Panchang Bedena, Batang Padang, Matang and Ampang Jajar Landfill. Air Hitam and Panchang Bedena Landfill gave exceeded amount of cadmium. The concentration of copper in Panchang Bedena, Batang Padang and Matang Landfill was recorded to exceed the standard limit.

It is interesting to note that the concentration of cadmium and zinc in the treated leachate collected at Air Hitam Landfill was found to be higher compared to the raw leachate in Table 2. This pattern was found to be similar as in JJLS, where the concentration of chromium was increased to a 51.63% after the raw leachate was subjected to treatment. Mat Salleh and Ku Hamid, 2013 explained that these occurrences were due to the accumulation of metals in the aeration pond and was collected together upon sampling. However, this phenomenon is related to the characteristics, distribution, and mobility of heavy metals in the landfill in a pH dependent and variable ratio of NH_3-N to total nitrogen content (Chai et al., 2007). The concentrations of heavy metals were found to be significantly higher in younger leachate (Renou et al., 2008). Low concentrations of heavy metals were expected for older, aged leachate in methanogenic phase and due to the decreased metal solubility in an alkaline condition (Renou et al., 2008; Kulikowska & Klimiuk, 2008). However, degradation of organic substances produces humic substances that could adsorb the heavy metals in the landfill that will elevate the concentration of heavy metals.

Table 2. Concentration of heavy metals in leachate in various landfills

Name of landfill/ metal	Jerangau-Jabor Landfill		Jeram Sanitary Landfill	Air Hitam Landfill		Panchang Bedena Landfill	Batang Padang Landfill	Matang Landfill	Ampang Jajar Landfill	Kulim Landfill	Kuala Sepetang Landfill	Acceptable conditions for the discharge of leachate
	Raw	Treated		Raw	Treated							
Ag	0.0041	0.0024	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.10
Cd	0.1078	0.0706	0.03	0.006	0.374	0.030	0.001	0.003	ND	ND	ND	0.01
Cr	0.4956	0.7515	0.65	0.003	0.002	0.030#	0.007#	0.020#	0	ND	ND	0.05# and 0.20
Cu	0.0333	0.0063	0.01	0.013	0.011	0.24	1.52	0.10	0	0.03	0.08	0.20
Fe	6.0606	4.3665	7.3	0.080	0.045	ND	ND	ND	3	0.38	2.18	5
Pb	0.0261	0.0107	0.03	0.017	0.00025	1.953	0.745	0.130	0.3	ND	ND	0.10
Zn	0.1882	0.0229	3.27	0.013	0.019	ND	ND	ND	0.01	0.09	0.26	2.0
References	This work		Jumaah et al., 2016	Mat Salleh & Ku Hamid, 2013		Nor Nazrieza et al., 2015			Umar et al., 2010	Zainol et al., 2012		Standard*

ND- not determined

*Environmental Quality (Control of Pollution From Solid Waste Transfer Station and Landfill) Regulations 2009 (PU(A) 433)

#concentration of hexavalent chromium

Table 3 exhibited the calculated sub-LPIhm for the landfills in Malaysia using the data from Table 2. The sub-leachate pollution index of heavy metals (sub-LPIhm) was one of the three sub-indices of LPI. The aggregation of these three sub-LPIs will result in the overall value of Leachate Pollution Index (LPI). The use of sub-LPIhm emphasized the strength of the selected heavy metals towards the level of contamination of leachate at a landfill.

The lowest calculated index value of

sub-LPIhm is 5.0 for JJLS and Air Hitam Landfill. Jeram landfill gave a sub-LPIhm of 5.202 followed by Ampang Jajar Landfill with the highest sub-LPIhm of 5.227. Although some of the heavy metals in JJLS have exceeded the standard but the value for the sub-leachate pollution index of heavy metals remains the lowest clearly showed that JJLS is a well sustainable landfill. It is noteworthy that the quantity of the generated leachate in landfills are extremely important and the sub-LPIhm is serve as a quantification of the level of heavy metal pollution and should always be presented along with the LPI value to give a larger description of the pollution potential level of the generated leachate. The sub-LPIhm can act as a preliminary hazard identification tool for heavy metal pollution in leachate.

Table 3. Sub-LPI for Jerangau-Jabor, Jeram, Ampang Jajar and Air Hitam Landfill

Parameter	**Variable weights**	**Sub-index values**						**Overall pollutant rating**					
		Jerangau-Jabor Landfill		**Jeram Landfill**	**Ampang Jajar Landfill**	**Air Hitam Landfill**		**Jerangau-Jabor Landfill**		**Jeram Landfill**	**Ampang Jajar Landfill**	**Air Hitam Landfill**	
		Raw	**Treated**			**Raw**	**Treated**	**Raw**	**Treated**			**Raw**	**Treated**
Cu	0.05	5	5	5	5	5	5	0.25	0.25	0.25	0.25	0.25	0.25
Pb	0.063	5	5	5	6	5	5	0.315	0.315	1.7325	0.378	0.315	0.315
Zn	0.056	5	5	6	5	5	5	0.28	0.28	0.28	0.28	0.28	0.28
Cr	0.064	5	5	5	5	5	5	0.32	0.32	0.32	0.32	0.32	0.32
Fe	0.045	5	5	5	5	5	5	0.225	0.225	0.225	0.225	0.225	0.225
Total	0.278							1.39	1.39	2.8075	1.453	1.39	1.39
Sub-LPI								5	5	10.099	5.227	5	5

CONCLUSION

The concentration of cadmium, chromium and iron in JJLS were found to exceed the standard limits and therefore not an acceptable condition for the discharge of leachate. It was found that all the treated leachate contained lower concentration of heavy metals than the raw leachate, except for chromium, which the concentration was increased about 51.63% in the treated leachate. Left without further action, the discharge of the leachate will definitely raise concern on the environmental pollution and the well-being of the community. However, the sub-leachate pollution index of heavy metals (sub-LPIhm) at Jerangau-Jabor Landfill Site (JJLS) showed that the level of heavy metal pollution potential of the leachate is low and that JJLS is well sustainable. Although the sub-LPIhm served as a quantification of the level of heavy metal pollution and should always be presented along with the LPI value to give an overall view of the pollution potential level of the generated leachate. The sub-LPIhm can served as a preliminary indicator to assess and monitor the level of leachate pollution threat due to the leaching of heavy metals from landfills. It is also recommended that frequent analysis and monitoring of individual heavy metals must be made from time to time to ensure the consistency of discharge leachate complied with standard limit set by the Environmental Quality (Control of Pollution From Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 (Act 127) with other standards and by-laws adopted by the local authorities to maintain the sustainability of JJLS.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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