

Leachate characterizations and pollution indices of active and closed unlined landfills in Malaysia

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ARTICLE INFO

Keywords:

Leachate characterization
Unlined landfill
Active landfill
Closed landfill
Leachate pollution index (LPI)

ABSTRACT

Leachate is one of the most serious environmental hazards associated with landfills especially which are unlined and uncontrolled. Thus, this study aims to characterize and compare raw leachate from active landfills (Ulu Maasop and Kampung Keru) and closed landfill (Pajam), then subsequently quantify the potential leachate contamination from each landfill using Leachate Pollution Index (LPI). The results obtained from the current study were compared with data from previous publications and indicated higher physico-chemical characterizations, especially for BOD and COD, than that of the other similar landfill categories. Based on the current findings, the low ratio of biodegradability and slightly alkaline pH values in leachate samples indicated that the sites (both active and closed landfills) were characterized by methanogenic conditions. Moreover, heavy metal concentrations, Cr, Fe and Se levels were higher than the leachate discharge standards for all leachate sampling sites. The As, Cu, and Mn, also surpassed the leachate discharge standards for the active landfill sites tested in this study (Ulu Maasop and Kampung Keru landfills). Next, the LPI calculated for Ulu Maasop, Kampung Keru, Pajam (1 and 2) landfills were 15.28, 13.89, 12.91 and 11.51, respectively, all exceeding the LPI for discharge standard of 5.696. Based on the leachate characterizations and the LPI values, these landfills pose a significant threat to the environment through the dissipation of leachate to the surface and groundwater, especially with the presence of BOD, COD, As and Cr. Thus, remedial actions such as rehabilitation of such unlined landfills and post-closure monitoring are crucial for both active and closed landfills to ensure the generated leachate is stabilized. Such actions will reduce and control the threat to the environment.

1. Introduction

Solid waste prevention and management are one of the key features of a sustainable environment and development (Aja et al., 2014). Among the developing country in Asia, most municipal solid wastes are disposed on land which could cause significant environmental impacts and environmental degradation (Bhalla et al., 2013). For instance, there are about 230 landfills in Malaysia which are mostly old and uncontrolled landfills with varying sizes operating without any environmental protection such as appropriate bottom liners and leachate collection systems. Most of these landfills are not classified as sanitary landfills because there are no facilities to collect and treat the leachate as well as no infrastructure to capture the landfill gas. There are only a

few sanitary landfills in Malaysia with leachate treatment and gas exploitation facilities (Agamuthu, 2001; Alkassasbeh et al., 2009).

Untreated leachate is one of the major hazards generated from municipal solid waste landfills (Abu-Daibes et al., 2013; Bhalla et al., 2013). Many researchers agree that leachate from active and closed unlined landfills can be a significant threat to the environment and the ecosystem as it may contaminate groundwater and surface water due to the dissipation of leachate through the soil (Ashraf et al., 2013; Naveen et al., 2017). The problem is worsened when the leachate travels or is transferred to nearby farms and residential areas with the occurrence of seasonal rain (Al-Raisi et al., 2014).

Leachate is formed as a result of multiple chemical and biological reactions of solid waste within the landfill (Abu-Daibes et al., 2013).

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<https://doi.org/10.1016/j.enmm.2019.100232>

Received 1 February 2019; Received in revised form 9 April 2019; Accepted 24 May 2019

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Table 1

Compositions of leachate during acid and methanogenic phases (Ehrig.H.j, 1988; Kjeldsen et al., 2002).

Parameter	Units	Acid Phase		Methanogenic Phase	
		Range	Average	Range	Average
pH		4.5 – 7.5	6.1	7.5 - 9	8
Biochemical Oxygen Demand (BOD ₅)	mg/l	4000 - 40000	13000	20 - 550	180
Chemical Oxygen Demand (COD)	mg/l	6000 - 60000	22000	500 - 4500	3000
Calcium	mg/l	10 - 2500	1200	20 - 600	60
Magnesium	mg/l	50 - 1150	470	40 - 350	180
Iron	mg/l	20 - 2100	780	3 - 280	15
Manganese	mg/l	0.3 - 65	25	0.03 - 45	0.7
Zinc	mg/l	0.1 - 120	5	0.03 - 4	0.6

Characteristics of leachate vary based on the heterogeneous solid wastes and normally depend on the compositions of waste mixtures, geographical conditions, the age of landfills / age of deposited wastes. The composition and mineralisation of the leachate are influenced by the physicochemical environment and microbial activities in the transformation of organic and inorganic compounds (Naveen et al., 2017). Various heavy metals like, arsenic, lead, chromium, mercury and cadmium which are naturally found in the soil or in the waste materials, have been detected in the leachate of the active solid waste landfill (Talalaj and Biedka, 2016). Such leachate plume downgradient in the landfills also undergo different phases of decomposition (Table 1). Another aspect to consider during the decomposition phase is the state of these parameters in the leachate as the landfill stabilises. Several parameters change dramatically as the landfill stabilises. Nevertheless, several inorganic parameters such as cadmium, chromium, cobalt, copper, lead, nickel and zinc will not be affected by landfill stabilisation, thus the concentrations will not reduce significantly (Christensen et al., 2001). There is a strong relationship between the state of refuse decomposition with leachate characteristics (Kjeldsen et al., 2002). Thus, leachate characterization serves as a guideline for the implementation of an appropriate leachate treatment

procedure.

Remedial processes such as leachate collection, leachate treatment, and monitoring of landfills are complex, thus, is usually costly (Tyrrel et al., 2002; Youcai et al., 2000). Thus, a rapid assessment and cost-effective alternative for an easy comparison of the leachate contamination potential from each type of landfill as a comparative scale in terms of Leachate Pollution Index (LPI) has been developed (Kumar and Alappat, 2005a). LPI is a formulated using the Rand Corporation Delphi Technique (Kumar and Alappat, 2005a; Rafizul et al., 2012) which serves as a vital tool for policymakers and public about pollution threat from the landfill. It is an environmental index which acts as a quantitative and comparative measure to quantify pollution potential of leachate generated at the landfill site (Kale et al., 2010; Lothe and Sinha, 2017). Also, it serves as an information tool which allows authorities to decide on top priority landfills which require instantaneous attention for remediation works (Tamru and Chakma, 2015). The LPI is a single number ranging from 5 to 100 that expresses the overall leachate contamination potential of a landfill based on several leachate pollution parameters at a given time. It is an increasing scale index, wherein a higher value indicates a poor environmental condition (Agbozu et al., 2015).

In this study, three unlined landfill sites, two active dumpsites and one closed dumpsite in Negeri Sembilan were characterised and compared based on their leachate contamination potential affected by the compositions of leachate produced in those landfills. The sites were chosen based on the perception that leachate from those unlined landfills would ultimately leak, percolate and contaminate the groundwater. Hence, this analysis can be used as an indication to test for environmental pollution (Ogundiran and Afolabi, 2008). This study analyses and compares the results of leachate compositions in both active and closed landfills and were also characteristically compared with related data from the literature. It is essential to understand the complex nature of leachate to propose for proper treatment. Moreover, very few published data are available on the leachate pollution potential of landfill sites in Malaysia. The present study was carried out to serve as an additional reported data for future reference. This is because LPI identifications can be used as a tool for researchers and authorities



Fig. 1. Sampling locations for leachate.

Image source: (a) Jabatan Pengurusan Sisa Pepejal Negara, 2015.

Table 2
Characteristics of landfills.

Sites	Location	Status	Period of landfilling (year)	Leachate collection and treatment
UML	Rural	Active	36	No
KKL	Rural	Active	20	No
PL (1 & 2)	Urban	Closed	23	Yes

to identify landfills which need urgent attention.

2. Materials and methods

2.1. Description of study areas

Leachate samples were obtained from 3 landfills in the state of Negeri Sembilan, to evaluate contamination potential based on operational status of the landfills which are active and safely closed dumpsites. Fig. 1 illustrates the landfill sites in the state of Negeri Sembilan and the study areas. Landfill leachate sampling was carried in September to October 2017. The characteristics of the landfill sites are listed in Table 2.

The study area for active dumpsite is in Ulu Maasop landfill (UML) which is located in the Kuala Pilah District. The first UML cell was safely closed in the year 2013. However, the remaining cells of the landfill are still active where they accept wastes from sources within Kuala Pilah area. This dumping site (about 36 years in operation) receives roughly 60 tonnes of waste per day (JPSPN, 2015). Originally, it was an open dumpsite (Level 0), which later on was upgraded to a controlled dumpsite (Level 1). The leachate generated in the landfill does not only originate from the active waste cells, but also from the closed cell.

Kampung Keru landfill (KKL) was opened in 1988 and receives about 135 tonnes (JPSPN, 2015) of commercial and domestic wastes per day. The waste disposed at this site practices no segregation and no soil cover over the deposited waste. Both the UML and KKL dumpsites have no lining system at the bottom of the deposited waste, permitting the untreated leachate to percolate into the soil and flow naturally as runoff ending up in the nearby drainage around the landfill. Since both of the landfills were not equipped with leachate collection system, leachate samples were collected randomly from the leachate streams within and around the sites.

Thirdly, the Pajam landfill (PL1 and PL2) area measures about 279 000 m² and was an active open tipping site before it was closed due to several issues for instance, odour pollution issues (Sakawi et al., 2011), river water contamination at Sungai Pajam (Zaini et al., 2010), landfill fires and also located near residential area (JPSPN, 2015). This landfill, which started its operation in the year 1993 with the capacity of 380 tonnes per day, was then converted into a renewable energy park (solar farm) which was able to generate approximately 8 MW of electricity. Upon its closure, the landfill safe closure procedures were implemented which involved the landfill to be equipped with (i) leachate collection and treatment system, (ii) leachate, groundwater and gas monitoring wells as well as (iii) soil capping systems. Leachate samples collected from this landfill were divided into two categories. As for sample PL1, leachate was collected from the surface drain which was located around the waste cells which were still undergoing safe closure procedure (Phase 2). The leachate through the surface drain will be pumped to the leachate treatment plant. The leachate in this area has the risk to overflow to the nearby drainage system during certain occurrences which can cause point source pollution to the receiving water body. Whereby, sample PL2 was collected from raw leachate collection pond, which receives leachate from both Phase 1 (completed safe closure procedure) and Phase 2 cells.

2.2. Leachate sampling and preservation

All leachate samples were collected (with minimum of three samples for each sites) and stored in clean 2 litres HDPE bottles that were thoroughly rinsed with deionised water and rinsed again with leachate samples prior to collection. 250 ml of samples were stored separately (for heavy metal analysis) where nitric acid was added to these samples to bring the pH to 2.0, the standard unit to prevent the precipitation of the metals. The remainder of the samples were left unacidified. The samples were then transported to the laboratory in a cool box and stored at 4 °C until further analyses to minimize biological and chemical reaction. The collection, preservation and measurement of samples were conducted according to the Standard Methods for Examination of Water and Wastewater (American Public Health Association - APHA, 2005).

2.3. Analytical procedures

The pH, temperature, dissolved oxygen (DO), conductivity (EC), turbidity, and total dissolved solids (TDS) concentrations for leachate samples were measured using a portable multimeter probe (HORIBA U-50 Series Multi-Parameter Water Quality Meter) in the field. Five days biochemical oxygen demand test (BOD₅) and chemical oxygen demand (COD) were analysed according to the standard method (5210B and 5220C, respectively). Preserved leachate were filtered using a syringe filter of 0.45 µm pore size and were subjected to measurement using Inductively Coupled Plasma – Optical Emission Spectrometry (ICP Optima 7300DV, Perkin-Elmer Instruments, USA) in BIOREC, Faculty of Civil Engineering, UiTM. Compressed air, purified nitrogen and argon gas were used for ICP-OES operation. A calibration blank and calibration standard (Perkin-Elmer, multi-elements, 1000 mg/l) stock solutions were prepared for a three-point calibration. Measurement for leachate samples were taken in triplicates under specific wavelength. The elemental concentrations of metals in landfill leachate namely calcium (Ca), magnesium (Mg), arsenic (As), cadmium (Cd), copper (Cu), cobalt (Co), chromium (Cr), nickel (Ni), zinc (Zn), manganese (Mn), iron (Fe), lead (Pb), selenium (Se) and thallium (Tl) were analysed based on the standard method (American Public Health Association - APHA, 2005). Spectral determination of the metals by ICP-OES was performed by measuring absorbance at the maximum wavelength of 317.1 nm for Ca, 279.5 nm for Mg, 188.9 nm for As, 214.4 nm for Cd, 324.7 nm for Cu, 228.6 nm for Co, 205.5 nm for Cr, 231.6 nm for Ni, 202.5 nm for Zn, 257.6 nm for Mn, 238.2 nm for Fe, 220.3 nm for Pb, 196.0 nm for Se, and 401.9 nm for Tl. Data are expressed as means of minimum three (3) replicates (including different trials).

2.4. Calculation of leachate pollution index (LPI)

In this study, the (18) parameters used for the estimation of LPI were pH, TDS, BOD₅, COD, TKN, NH₄N, TCB, phenolic compounds, As, Cr, Cu, Fe, Ni, Zn, Pb, Hg, cyanide, and chlorides. The LPI was calculated according to Eq. (1) as follows (Kumar and Alappat, 2005a):

Leachate Pollution Index (LPI)(when all 18 parameters are available)

$$= \sum_{i=1}^n w_i p_i \quad (1)$$

Where n is the number of leachate pollutant parameters, w_i is the weight for the i th pollutant variable and p_i is the sub-index values of the i th leachate pollutant variable. However, this particular equation is used when all eighteen (18) selected variables (Table 2) are known ($n = 18$ and $\sum w_i = 1$). Nevertheless, in this case, since not all pollutant variables were known ($m < 18$ and $\sum w_i < 1$), Eq. (2) was used.

Table 3

Comparison of leachate characteristics from this study with those of other active landfills in Malaysia.

Parameters	Standard*	Ulu Maasop, UML	Kg. Keru, KKL	Kulim	Kuala Sepetang	Kulim	Kuala Sepetang	Matang	Kuala Sembeling	Matang	Panchang Bedena	Batang Padang	Bukit Beruntung
		This study	This study	(Aziz et al., 2010a,b)	(Aziz et al., 2010a,b)	(Zainol et al., 2012)	(Zainol et al., 2012)	(Zin et al., 2013)	(Zin et al., 2013)	(Nor Nazrieza et al., 2015)	(Nor Nazrieza et al., 2015)	(Nor Nazrieza et al., 2015)	(Jayanthi et al., 2016)
Age of deposited wastes		36	20	14	12	16	14	15	23	17	32	25	14
pH Value	6.0–9.0	7.76	8.59	7.8	8.1	7.59	8.05	7.6	8	6.76	8.6	7.67	7.09
Temperature	40	33.9	29.9							29.47	29.3	33.02	
TDS		11400	6810	6900	12568								
DO		2.3	1.91										
Turbidity		> 1000	> 1000			26	88.9						
Conductivity		23	13.57			2.92	11.9						
BOD ₅ at 20 °C	20	614	610	515	85	29	158	146	61	100.29	106.7	144.07	259
COD	400	7624	5082	1593	990	117	855	828	363	257.45	153.7	1481	985
BOD ₅ /COD ratio		0.081	0.12	0.323	0.086	0.248	0.185	0.176	0.168	0.39	0.694	0.097	0.263
Calcium, Ca		67.78	127.1										91.2
Magnesium, Mg		42.17	30.85										96.6
Arsenic, As	0.05	0.274	0.068										
Cadmium, Cd	0.01	0.005	0.003					0.19	0.14	0.003	0.03	0.001	0.4
Copper, Cu	0.2	0.21	0.084	0.2	0.7	0.03	0.08	0.09	0.1	0.1	0.24	1.52	2.62
Cobalt, Co		0.025	BDL										
Chromium, Cr	0.05	0.174	0.1	0.1	0.05								17.3
Nickel, Ni	0.2	0.116	0.114	0.2	0.3	0.07	0.16	0.17	0.1				12
Zinc, Zn	2	0.652	0.656	0.3	0.2	0.09	0.26						236
Manganese, Mn	0.2	0.312	0.304			0.09	0.08	0.09	0.22				5.1
Iron, Fe	5	14.56	16.594	6	5	0.38	2.18	2.3	8.3				60
Plumbum, Pb	0.1	BDL	0.012	1.6	0.4					0.13	1.953	0.745	1.12
Selenium, Se	0.02	0.251	0.001										
Thallium, Tl		0.257	0.02										

All units in mg/l except for pH, BOD/COD ratio, temperature (°C) turbidity (NTU) and conductivity (ms/cm); Standard - Acceptable Conditions for Discharge of Leachate, Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfills) Regulations 2009; BDL means below detection limit; Age of deposited wastes are considered based on the year of leachate sampling; Data are based on field work done on September – October 2017; Obtained results for this study were the mean value of minimum three determinations carried out simultaneously, (n = 4) leachate samples for UML and (n = 3) for KKL.

Leachate Pollution Index (LPI)(when NOT ALL 18 parameters are

$$available) = \frac{\sum_{i=1}^m w_i P_i}{\sum_{i=1}^m w_i} \quad (2)$$

Pollutant weight or weight factor, w , indicates the significance level of each pollutant to the overall leachate pollution. The weight factors for each pollutant variable in this study are summarised in Appendix 1 (Kumar and Alappat, 2005b). Next, individual pollution rating or the sub-index value, p , was obtained by referring to sub-index average curves of pollutants as shown in Appendix 2 (Kumar and Alappat, 2005b). The p values obtained for the parameters were multiplied with the respective weight factors, w . The weighted sum of all the parameters indicates the overall leachate pollution index (LPI) for each landfill.

3. Results and discussion

3.1. Characterizations of leachate

Tables 3 and 4 summarise the characteristics of raw leachate from the active and closed landfill sites, which are UML, KKL PL1 and PL2. The analysis was conducted in triplicates in which the data presented is the average for each sample. A comparison between these data with those obtained from other landfill sites (actives and closed landfills) are presented in the same table (Tables 3 and 4).

3.1.1. pH, Temperature and Dissolved Oxygen (DO)

The pH values recorded for leachate from all sampling locations were within the range of 7.5 to 9, which were in the alkaline range. The

pH observed in this study agrees with those in the previous studies, based on the categories and age of deposited wastes (Ashraf et al., 2013; Atta et al., 2015; Aziz et al., 2010a, b; Emenike et al., 2012; Jayanthi et al., 2016; Nor Nazrieza et al., 2015; Zainol et al., 2012; Zin et al., 2013). The alkaline nature of leachates indicates the mature stage of the dumping site (Jorstad et al., 2004). The pH of leachate becomes alkaline in nature due to the decrease in the concentration of partially ionized free volatile fatty acids which are being used up by the methane-producing bacteria. Furthermore, pH of leachate tends to increase gradually with time from slightly acidic towards alkaline values as the site gets older and more stabilized. UML, KKL and PL are considered as old landfills, since they have been under operation for 36, 20 and 23 years, respectively. The long years of operation of these landfills strongly agree with the evidence of leachate's higher pH values (> 7.5) for old landfills (Abbas et al., 2009) where they are also capable of carrying a greater load of dissolved substances (Naveen et al., 2017). Therefore, even with the continuation of new waste deposition at these landfill sites (active landfills), the acidogenic leachates was not observed as the ratio of the old and stabilized waste to the newly deposited waste was high and so was the alkalinity (Demirbilek et al., 2013).

In addition, the temperature of leachate is also an important factor which increases the biological activities and decreases the DO amount (Demirbilek et al., 2013). Based on the observation, the recorded temperatures did not vary significantly among all sampling sites. The concentrations of DO is an indicator of the distribution of flora and fauna (Naveen et al., 2017). Hence, reduction in DO levels may sometimes cause changes in biological diversity. Leachate from municipal landfill usually contains very low DO levels due to waste compression

Table 4

Comparison of leachate characteristics from this study with those of other closed landfills in Malaysia.

Parameters	Standard*	Pajam, PL1	Pajam, PL2	Ampang Jajar (T.S)	Air Hitam	Air Hitam	Ampar Tenang	Taman Beringin (T.S.)	Taman Beringin (T.S.)
		This study	This study	(Aziz et al., 2010a)	(Emenike et al., 2012)	(Nur Fatin Dahlia & Ku Halim, 2013)	(Ashraf et al., 2013)	(Atta et al., 2015)	(Jayanthi et al., 2016)
Age of deposited wastes		23	23		17	15	12	21	24
pH Value	6.0–9.0	8.74	7.88	7.5	8.2	6.96–8.49	8.10–8.24	7.8	7.57
Temperature	40	29.11	30.62		29.5	26.44–26.68	28.44–29.40		
TDS		11100	7290	2543	830		3876–3989		
DO		0.9	2.48		5.8		0.12–0.18		
Turbidity		177	216		108				
Conductivity		17.9	11.8		20		8.10–8.24		
BOD ₅ at 20°C	20	370	322	48	3500		256–288	90.5	127
COD	400	3953	2880	599	10234	1239–3607	3187–3222	456.16	482
BOD ₅ /COD ratio		0.094	0.112	0.08	0.342			0.198	0.263
Calcium, Ca		102.7	92.38		25.6	0.949–1.059	9.1–9.9		242.1
Magnesium, Mg		33.43	27.14		20.3	0.314–0.416	52.23–53.19		52.2
Arsenic, As	0.05	0.007	0.003			0.011–0.232	0.210–0.230	0.08	
Cadmium, Cd	0.01	0.008	0.007		< 0.001	0.006–0.374	0.089–0.098	0.018	0.4
Copper, Cu	0.2	0.021	0.019	0	< 0.001	0.011–0.013	0.770–0.790	0.041	0.5
Cobalt, Co		0.016	0.013						
Chromium, Cr	0.05	0.109	0.061	0	0.11	0.002–0.004	0.012–0.022	0.03	6.2
Nickel, Ni	0.2	0.118	0.065	0	0.29	< 0.200	0.789–0.817	0.031	0.85
Zinc, Zn	2	0.157	0.116	0.01	0.1	0.013–0.032	0.642–0.666	0.17	24.3
Manganese, Mn	0.2	0.129	0.174		0.12	0.005–0.011	0.080–0.100	0.27	3.1
Iron, Fe	5	5.868	7.854	3	3.1	0.080–0.159	2.180–2.910	4.78	134.6
Plumbum, Pb	0.1	0.014	0.012	0.3	< 0.001	0.004–0.017	0.230–0.240	0.07	< 0.01
Selenium, Se	0.02	0.032	0.044						
Thallium, Tl		0.199	0.223						

All units in mg/l except for pH, BOD/COD ratio, temperature (°C) turbidity (NTU) and conductivity (ms/cm); Standard - Acceptable Conditions for Discharge of Leachate, Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfills) Regulations 2009; BDL means below detection limit; Age of deposited wastes are considered based on the year of leachate sampling; Data are based on field work done on September – October 2017; Obtained results for this study were the mean value of minimum three determinations carried out simultaneously, (n = 3) leachate samples for each PL1 and PL2.

processes and aerobic decomposition of the wastes in which microbes use up oxygen to transform organic materials to inorganic substances (Fetter, 2001). The DO concentrations measured in this study for both the active and closed landfills exhibited slightly low value, with the minimum and maximum concentrations of 0.91 and 2.48 mg/l, respectively. Ashraf et al. (2013) conducted a similar study in Ampar Tenang Landfill also reported even lower values of DO in leachate (0.12 to 0.18 mg/l) compared to the current study.

3.1.2. Total dissolved solids (TDS), conductivity (EC) and turbidity

EC is sensitive towards the variations in TDS, which postulates a strong correlation between these parameters (A. Al-Yaqout and Hamoda, 2003; Fan et al., 2006). The highest EC was reported for UML (23 ms/cm) with corresponding high TDS value (11400 mg/l). The TDS values in active landfills were observed to be higher than in closed landfills in this study and other previously cited studies, except for PLI which contained relatively higher TDS and EC. High TDS and EC may indicate the presence of dissolved organic and inorganic substances in the samples (A. Al-Yaqout et al., 2005; Kurniawan et al., 2006; Naveen et al., 2017) and high suspended matter (Ishak et al., 2016) and turbidity. The amount of TDS reflects the extent of mineralization, as a higher concentration of TDS can change the physical and chemical characteristics of the receiving water (Aziz et al., 2010a, b). Thus, relatively high TDS and turbidity values observed in all sites may lead to a reduction of water clarity, hence, contributes to a light limitation resulting in a decrease in photosynthesis. High TDS may also limit the growth and may consequently lead to the death of many organisms of the receiver water bodies (Naveen et al., 2017). Turbidity recorded in active landfills (UML and KKL) was significantly higher (> 1000 NTU) compared to closed landfills. Nevertheless, the observed results of high turbidity were not consistent with those in the previous studies with active landfills (Mohd Zin et al., 2012; Zainol et al., 2012), except for Kulim anaerobic landfill leachate which reported a maximum value of

4500 NTU (Aziz et al., 2010a, b). High turbidity may also be due to the nature of anaerobic leachate and may indicate the presence of high organic matter with some present in soluble form (Hamidi et al., 2007; Nor Nazrieza et al., 2015).

3.1.3. Biochemical oxygen demand (BOD₅) and chemical oxygen demand (COD)

Measurement of organic materials such as BOD₅ and COD is important to identify the strength of the leachate produced in landfills. BOD₅ is the evaluation of the amount of organic pollutant in water and wastewater, which is basically determined by measuring the DO which is being used up by microorganisms during the biochemical oxidation of organic matters (Metcalf and Eddy, 2003). COD, on the other hand, measures the oxygen required for organic waste constituents to completely oxidize into inorganic end products (Bhalla et al., 2012) and estimates the presence of toxic chemicals as well as oxidisable pollutants (Enitan et al., 2018).

In this study, the concentrations for BOD₅ and COD in both active and closed landfills were found to be very much higher than the effluent standard limit of leachate in Malaysia, which is similar to other landfills as cited in this study. Measured BOD₅ and COD values also surpassed the permissible limit of leachate discharge as reported in previous studies (Abu-Daibes et al., 2013; a. F. Al-Yaqout et al., 2005; Fan et al., 2006; Naveen et al., 2017; Xaypanya et al., 2018). The BOD₅ and COD concentrations in UML and KKL (active landfills) were found to be twice as higher than the leachate samples observed in PL (closed landfill). This result indicated that the organic contents in closed landfills are significantly lesser than that of active landfills. To the contrary, Emenike et al. (2012) reported significantly higher BOD₅ and COD concentrations (3500 and 10,234 mg/l respectively) at the Air Hitam closed landfill, suggesting that the wastes at closed landfills can remain completely undegraded and thus, possess high organic strength. Although the three dumpsites are old landfills, high BOD₅ and COD in

UML and KKL (active landfills) indicated that the waste deposited at both landfills has not yet stabilized.

On the other hand, the leachate in all sampling sites has shown the characteristics of methanogenic quality (Christensen et al., 2001). During the methanogenic phase, pH value will normally increase (from an average of 6.1 to the range of 7.5–9), while the concentrations of BOD₅ and COD, as well as the BOD₅/COD ratio will decrease (Christensen et al., 2001; Ehrig, 1988). Freshly deposited wastes will normally lead to higher degradability of organic compounds (acid phase) with significantly high BOD₅ and COD concentrations (Tatsi and Zouboulis, 2002a). Nevertheless, in this study, even with continuous new deposition of wastes into the active landfills (UML and KKL), the characteristics of the acidogenic phase was not observed. In this state, the expanding acid phase is suppressed by the dominant characteristics of the methanogenic phase, which probably due to high organic acid degradation (Erses et al., 2008) or higher ratio of old and stabilized waste compared to the newly deposited waste (Demirbilek et al., 2013).

3.1.4. BOD₅/COD ratio

Generally, the proportions of biodegradable organic constituents in landfill leachate is denoted by BOD₅/COD ratio. BOD₅/COD ratio also indicates the maturity of the landfills (El-Fadel et al., 2002). A ratio greater than 0.4 denotes leachate in the acid phase, while a ratio below 0.1 indicates low biodegradability of organic contents in the leachate (Ehrig, 1988). According to Lo (1996), a decrease in BOD₅/COD ratio can be observed as the landfills / wastes ages. This phenomenon occurs when much of the biodegradable organic material can easily be removed during the early stages of landfilling through biological processes. In the current study, the BOD₅/COD ratios for the collected leachate samples were 0.081, 0.12, 0.094 and 0.112 in UML, KKL, PL1 and PL2, respectively. Lower BOD₅/COD ratios (≤ 0.1) observed indicated that these landfills had reached a stable status due to the age of landfills (> 20 years), which can be considered as old landfills and might contain a considerable amount of biologically inert materials (Kurniawan et al., 2006; Sewwandi et al., 2013). A similar ratio was also observed in Batang Padang (Nor Nazrieza et al., 2015) and Ampang Jajar landfills (Aziz et al., 2010a, b). Leachate with low BOD₅/COD ratios is more suitable to be treated with physicochemical treatment techniques rather than biological method (Diamadopoulos, 1994; Kurniawan et al., 2006) due to the higher concentration of non-biodegradable organic compounds (Ntampou et al., 2006). However, higher BOD₅/COD (> 0.1) observed in several medium to young landfills cited in this study (< 20 years of landfilling) such as in Kulim, Matang and Panchang Bedena, Bukit Beruntung, and Taman Beringin landfills suggested that the organic materials found in the leachate are biodegradable, hence biological treatment process is more suitable.

3.1.5. Inorganic constituents

According to (Lo, 1996) and (Tatsi and Zouboulis, 2002b), fresh leachate samples from young landfills in Hong Kong and in Thessaloniki, Greece contained high concentrations of heavy metals. However, the inorganic concentrations tend to decrease as the landfill age increases mainly due to lesser metal solubilisation (caused by the increase in pH), adsorption and precipitation reactions of stabilized leachate. The availability and levels of heavy metals in landfills, especially for unlined disposal sites should be monitored to prevent contamination of surrounding soil and groundwater.

In this study, leachate from the active landfills exhibited slightly higher levels of inorganic constituents, especially As, Cu, Cr, Mn, Fe and Se, which is mainly associated with the active status (operational) of UML and KKL due to continuous deposition of fresh waste. As concentration measured in UML leachate was 0.274 mg/l, more than five times higher than the acceptable limit for discharge of leachate. High values of As was also observed in some closed landfills as reported by (Nur Fatin Dahlia and Ku Halim, 2013) in Air Hitam landfill (0.232 mg/l), Ashraf et al. (2013) in Ampar Tenang landfill (0.230 mg/l) and Atta

et al. (2015) in Taman Beringin landfill (0.080 mg/l). As normally comes from uncontrolled disposal of various electronic wastes, for instance circuits' boards, computer chips and LCD displays. Besides that, fertilizers have also been reported as a source of As contaminations (Abu-Daibes et al., 2013). As is a great pollutant concern not only due to the level of toxicity, but it is also has high solubility in water and not easy to remove. Apart from As, Cr and Fe were also found in high concentrations in all sampling locations for both active and closed landfills. High concentrations of Fe in all three landfills could be due to the dumping of metal scrap and tin-based garbage (Kale et al., 2010), which may also contribute to the high As concentration in UML. High concentrations of Cr revealed the presence of wood preservatives and paint products in the waste (Kale et al., 2010) and also from electroplating, spent rechargeable and household batteries, and tannery industry (Abu-Daibes et al., 2013). Moreover, discarded plastic materials and coloured polythene bags, as well as empty paint container might also be the source for Cr contaminations in landfill leachate (Parth et al., 2011). Objects like fluorescents lamps, refused batteries and other metallic items were also the main contributors to the rise of inorganic constituents in landfills (Mor et al., 2006). Other metals, Cu, Ni, Zn and Pb in all samples remain within the allowable limits for leachate discharge.

3.2. Leachate Pollution Index (LPI)

LPI provides a meaningful method of evaluating the contamination potential of different landfill sites at a particular time (Rafizul et al., 2011). Tables 5 and 6 illustrates the leachate contamination potential in terms of pollution rating (LPI) for UML, KKL and PL landfills and also compared to LPI for leachate discharge standard (Acceptable Conditions for Discharge of Leachate, Environmental Quality Regulation 2009). LPI was calculated on the basis of the available data, since not all the data for the parameters included in the LPI were available. Nevertheless, it is worth noting that each parameter in leachate properties has a significant impact on the LPI calculations (Mor et al., 2018). The site-specific comparison between the landfills (active and closed landfill sites) is illustrated in Fig. 2.

Based on the evaluated results, calculated LPI were 15.28 (UML) and 13.89 (KKL) for active landfill sites and 12.91 (PL1) and 11.51 (PL2) for a closed landfill site. The LPI for the active landfills were relatively higher than that of the closed sites. Nevertheless, the LPI values for all of the dumpsites investigated were above the LPI for standard leachate discharge (5.696). This is an indication that the leachate from each dumpsite has the capacity to contaminate the groundwater within the vicinity of the landfill (Ofomola et al., 2017) especially due to the absence of proper liner system underneath the deposited waste in all the dumpsites. However, the risk is higher especially for both of the active dumpsites as the leachate is not collected for treatment and can easily percolate to the surrounding area through the soil, and leach to the groundwater or to nearby waterways. Regrettably, a few studies conducted in Malaysia revealed that the groundwater and soil in the vicinity of disposal sites have already been contaminated (Ashraf et al., 2013; Mohd Raihan Taha et al., 2011; Norkhadajah et al., 2015; Nur et al., 2013; Rahim et al., 2010; Zaini Sakawi et al., 2013; Samudung et al., 2012, 2009; Tadza et al., 2000; I Yusoff et al., 2008; Ismail Yusoff et al., 2013). Moreover, as reported by Siti Nur Syahirah et al. (2013), the soil in Ampar Tenang (which is a closed landfill) was no longer capable of preventing pollution migration due to leachate seepage through the soil.

The high values of LPI in UML and KKL (active dumpsites) are attributable to high concentrations of BOD, COD and certain metal elements (As, Cr and Fe). Both active dumpsites have slightly higher LPI values as compared to the closed dumpsite, considering the disposal sites are still in operation and are receiving domestic, commercial, agricultural and might also receive industrial wastes as well. Aziz et al. (2010a, b) calculated LPI values for two active and one closed dump

Table 5
Leachate pollution index (LPI) for active landfill sites.

Parameters	Pollutant concentrations, c_i		Sub-index value, p_i		Pollutant weight, w_i		Overall pollution rating, $p_i w_i$		Standard pollution rating
	UML	KKL	UML	KKL	UML	KKL	UML	KKL	Standard
pH Value	7.76	8.59	5	5	0.055	0.055	0.275	0.275	0.275
Total Dissolved Solids (TDS)	11400	6810	25	15	0.050	0.050	1.250	0.750	–
Biochemical Oxygen Demand (BOD ₅) at 20 °C	614	610	24	23	0.061	0.061	1.464	1.403	0.366
Chemical Oxygen Demand (COD)	7624	5082	72	68	0.062	0.062	4.464	4.216	0.930
Arsenic, As	0.274	0.068	5	5	0.061	0.061	0.305	0.305	0.305
Copper, Cu	0.210	0.084	6	5	0.050	0.050	0.300	0.250	0.250
Chromium, Cr	0.174	0.100	5	5	0.064	0.064	0.320	0.320	0.320
Nickel, Ni	0.116	0.114	5	5	0.052	0.052	0.260	0.260	0.260
Zinc, Zn	0.652	0.656	5	5	0.056	0.056	0.280	0.280	0.280
Iron, Fe	14.560	16.594	5	5	0.045	0.045	0.225	0.225	0.225
Plumbum, Pb	0.000	0.012	5	5	0.063	0.063	0.315	0.315	0.315
Total					0.619	0.619	9.458	8.599	3.526
LPI values							15.28	13.89	5.696

Standard - Acceptable Conditions for Discharge of Leachate, Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfills) Regulations 2009; All units for pollutant concentrations are in mg/l except for pH, temperature (°C), turbidity (NTU) and conductivity (mS/cm); UML – Ulu Maasop Landfill; KKL – Kampung Keru Landfill.

sites in Malaysia. They reported the LPI values of 19.50 and 21.77 for the two active dumpsites and 16.44 for closed dumpsite. Their LPI values were higher, compared to the values observed in this study. Ofomola et al. (2017) and Salami et al. (2015) also calculated values for active dumpsites in Ughelli and Lagos, Nigeria. Their values were within the range of the results of this study, compared to the LPI values observed by Kumar and Alappat (2005c) which were 36.48 and 39.04 for active dumpsites and 45.01 and 15.97 for closed dumpsites. The LPI values from the latter are very higher, due to the relatively higher concentrations of BOD and COD in the leachate. The similarity among other studies, Aziz et al. (2010a, b) for landfills in Malaysia, Kumar & Alappat (2005c) for landfills in Hong Kong, and De et al. (2016) in their studies in Kolkata, India, with this study is that the LPI values for closed dumpsites (except Ma Tso Lung landfill) were observed to be lower than that of the values in active dumpsites. This indicates that the closed landfills have stabilised thus, contamination potential is reduced. As for the condition of Ma Tso Lung landfill (Kumar and Alappat, 2005b) which recorded high LPI value despite the inactive landfill status, the leachate produced by this closed landfill might still be hazardous and requires appropriate post-closure monitoring.

Table 6
Leachate pollution index (LPI) for closed landfill sites.

Parameters	Pollutant Concentrations, c_i		Sub-index value, p_i		Pollutant weight, w_i		Overall pollution rating, $p_i w_i$		Standard pollution rating
	PL1	PL2	PL1	PL2	PL1	PL2	PL1	PL2	Standard
pH Value	8.74	7.88	5	5	0.055	0.055	0.275	0.275	0.275
Total Dissolved Solids (TDS)	11100	7290	25	15	0.050	0.050	1.250	0.750	–
Biochemical Oxygen Demand (BOD ₅) at 20 °C	370	322	13	12	0.061	0.061	0.793	0.732	0.366
Chemical Oxygen Demand (COD)	3953	2880	60	55	0.062	0.062	3.720	3.410	0.930
Arsenic, As	0.007	0.003	5	5	0.061	0.061	0.305	0.305	0.305
Copper, Cu	0.021	0.019	5	5	0.050	0.050	0.250	0.250	0.250
Chromium, Cr	0.109	0.061	5	5	0.064	0.064	0.320	0.320	0.320
Nickel, Ni	0.118	0.065	5	5	0.052	0.052	0.260	0.260	0.260
Zinc, Zn	0.157	0.116	5	5	0.056	0.056	0.280	0.280	0.280
Iron, Fe	5.868	7.854	5	5	0.045	0.045	0.225	0.225	0.225
Plumbum, Pb	0.014	0.012	5	5	0.063	0.063	0.315	0.315	0.315
Total					0.619	0.619	7.993	7.122	3.526
LPI values							12.91	11.51	5.696

Standard - Acceptable Conditions for Discharge of Leachate, Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfills) Regulations 2009; All units for pollutant concentrations are in mg/l except for pH, temperature (°C), turbidity (NTU) and conductivity (mS/cm); PL1 – Pajam Landfill 1; PL2 – Pajam Landfill 2.

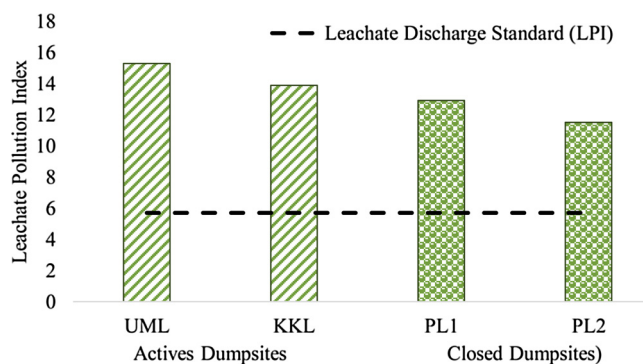


Fig. 2. Variations in leachate pollution index.

4. Conclusions

The present study investigated the characteristics of leachate emerging from active and closed unlined landfills in Malaysia. The

leachate derived from all three dumpsites in this study demonstrated relatively high values of BOD, COD, and potentially toxic heavy metals (As and Cr). It is important to realize that As and Cr are considered to be dangerous pollutants, and are toxic even at lower concentrations. Based on the characterization of leachate samples, both active and closed landfills are in the stable methanogenic phase which might possibly due to the age of deposited wastes. Higher pH values, BOD, COD concentrations and low BOD₅/COD ratios were also suggestive of the methanogenic phase of the leachates. In this phase, physicochemical treatment such as adsorption and reverse osmosis will be more effective compared to biological treatments. Chemically-aided post-treatment system was also recommended for older leachate or leachate having low BOD₅/COD ratios. Furthermore, a relatively low organic and inorganic strength of leachate produced in a closed landfill compared to the active landfills might be due to the minimal substrate present for microbiological activities.

LPI is a reliable hazard identification tool for the policymakers and the public regarding the leachate pollution threat from the landfills. Between the active and closed dumpsites considered for evaluating leachate contamination potential, the high value of LPI values from active dumpsites (UML and KKL) indicated that the leachate should be prioritised for immediate attention. Moreover, the leachate from UML possessed the highest risk of environmental pollution based on the characterization of leachate parameters and also calculated LPI values. It can also be concluded that concentrations of BOD, COD and certain metal elements (As, Cr and Fe) are attributable to the high values of LPI. Though these landfills are located in the rural areas and are predicted to receive only domestics and commercial wastes, the exceedance in certain highly toxic metals such as As, and Cr may suggest the presence of illegal dumping of hazardous wastes or lack of proper segregation of wastes before dumping in the landfill. On the other hand, contamination potential from closed landfill (LP1 and LP2) was slightly lower, but still requires appropriate post-closure monitoring. In a nutshell, proper remediation is crucial for both active and closed landfills knowing that the landfills were unlined, to avoid further spreading of contaminated leachate to the environment.

5. Recommendations

Though remedial measures cannot be undertaken in one go due to financial constraints, appropriate preventive measures should be implemented immediately to reduce the impact on land and surface water contamination especially from unlined dumpsites. The preventive and mitigation measures proposed are as follows:

- 1 Developing low-cost and site-specific leachate treatment facilities;
- 2 In the case of unlined landfills, leachate generated should be collected by constructing boreholes where the leachate should be diverted to a pool (with proper lining system) for further treatment;
- 3 Proper segregation of biodegradable, non-biodegradable and recyclable (especially plastic and metals to reduce inorganic and heavy metal loads through the leachate);
- 4 Continuous disposal of wastes at uncontrolled dumping sites should be discouraged;
- 5 Proper evacuation and clean-up program should be conducted;
- 6 Rehabilitation of old unlined landfills with continuous groundwater monitoring programmes;
- 7 Active post-closure (for closed dumpsites) monitoring is required until the leachate generation is stabilized and poses no further threat to the environment.

Conflict of interest and authorship conformation form

Please check the following as appropriate:

- All authors have participated in (a) conception and design, or

analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.

- This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue.
- The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.

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Acknowledgments

The authors would like to express appreciation for the support of the study provided by Universiti Teknologi Malaysia, through Tier 1 Grant [Project Number = Q.K130000.2543.15H73]. This study is also supported by the Takahashi Industrial and Economic Research Foundation Grant, Japan.

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