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ORIGINAL ARTICLE

The potential impact of leachate-contaminated groundwater of an ex-landfill site at Taman Beringin Kuala Lumpur, Malaysia

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Abstract Spreading of leachates to soil, surface and groundwater aquifers is a prevailing problem with unlined landfills, causing many problems with adverse impacts on the surrounding environment. The impact of leachate contamination on groundwater quality was investigated from unlined ex-landfill site at Taman Beringin in Malaysia to identify the characteristics of the groundwater, and the possible impacts on the environment. Various physico-chemical parameters including: temperature, pH, color, TDS, TSS, BOD, COD, S^{2-}, F^-, Cl^-, NH_3-N, SO_4^{2-}, NO_3^-, and heavy metals: Ag, As, Cd, Cu, Cr^{3+}, Cr^{6+}, Fe, Hg, Ni, Pb, Zn, Mn, B, Se, Sn and Ba, were collected from four (4) monitoring wells and six (6) surface raw leachate points between December 2012 and January 2013. Physico-chemical analyses of sampled leachates and groundwater followed standard analytical methods. The results showed considerable impact of leachates on groundwater with high concentration of most chemical parameters in groundwater samples: COD (101.75 ± 99.42), BOD (28 ± 30.99), NH_3-N (31.10 ± 19.12), F^- (0.72 ± 0.32) and heavy metal; Pb (0.042 ± 0.045), Ni (0.016 ± 0.006), Fe (0.41 ± 0.68) have exceeded the Malaysian National Drinking Water Quality Standard and National Water Quality Standards CLASS II A. Remedial measures are suggested to prevent further spreading of leachates into rivers via groundwater flow. In addition, combined process of routine chemical treatment prior to biological treatment is necessary to improve the existing leachate quality to minimize the effects on the surrounding environment.

Keywords Landfill leachate · Biological oxygen demand (BOD) · Ammonical nitrogen · Groundwater contamination · Water Quality Standards

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Introduction

The degradation of waste into constituent chemicals occurs in stages in landfills and this may continue for several years even long after their closure, while they continue to produce leachates and landfill gases. Spreading of leachates is however, a phenomenon that is evident around landfill without proper lining material or when lining material is punctured or leaky (Agamuthu 2001). However, some contaminants concentrations in the leachates could be present at concentrations above the appropriate standards, while some could stay high for extended period of time. These contaminants threaten groundwater resources and consequently, can pose a serious threat to human, animal and aquatic life and the deterioration of the ecosystems as a whole.

Landfill design has often been one of the means of the source of groundwater pollution in Malaysia. The problems have also been aggravated from the fact that most of the existing solid waste disposal sites are practicing either open dumping or controlled tipping because the technology of proper landfill operation is not properly implemented (Chong et al. 2004). In accordance with the Eighth Malaysia Plan (2001–2005), the Department of

Environment Malaysia (DOE) enlisted the rehabilitation of 26 polluted rivers. Three of the river basins studied were in Peninsular Malaysia and are under the DOE scheme of river rehabilitation. These include: Langat River in Selangor, Tebrau and Segget in Johor (Mamun and Zainudin 2013). The study revealed that the frequent parameters which have frequently exceeded the NWQS Class II A limits include: BOD, COD (chemical oxygen demand), ammoniacal nitrogen (NH_3-N), and suspended solids (SS) (Mamun and Zainudin 2013). In addition, the results obtained by Juahir et al. 2010; Othman et al. 2001; Hoo et al. 2005, showed that NH_3-N, nitrate (NO_3), BOD, COD, SS and heavy metals are associated mainly due to anthropogenic sources and are the main contaminant in the Langat River Basin. These chemical substances make up the frequent and potential contaminant of soil, surface and groundwater around landfill and the main pollutants to render many of Malaysian rivers polluted. Landfill leachates wastewater, however, contributes to the main point sources responsible for the situations of degraded river water in Malaysia and their potential impact on groundwater resources cannot be underestimated. Elevated level of NH_3-N had resulted in the temporary closing of some landfills such as the Cheras WTP, thrice in 1997 (Mamun and Zainudin 2013).

Delphi approach was carried out to assess seven selected disposal sites out of the ten waste disposal sites used to receive waste in the Federal Capital Territory of Kuala Lumpur, Malaysia. Taman Beringin landfill (TBL) ranked highest of the most polluted landfill with the Landfill Pollution Index (LPI) of 719.56 (Chong et al. 2001, 2004). TBL site was also the worst in terms of overall environmental conditions which had taken into consideration nine different criteria including: gas emissions, chemical constituents and water quality with 59 selected parameters showing that the level of pollution assessed at the site was much exceeded the threshold limits of 179.58 (Chong et al. 2001, 2004).

The Taman Beringin ex-landfill is owned and operated by Kuala Lumpur City Hall (KLCH), also known as Dewan Bandaraya Kuala Lumpur (DBKL). The landfill had begun operating since 1991. At the time of operation, due to the rapid growth in the socio-economic development of Kuala Lumpur, TBL exceeded its operational lifespan. Robinson and Carville (2010) pointed out that the landfill comprised in excess of 20 million tons of domestic wastes. Moreover, environmental degradation to the residents and surrounding areas caused by the landfill operations resulted in leachate contamination of surface/ground water, landfill gas migration, odor and vector-borne nuisances, vermin, fires and extensive water pollution (CyEn 2005; Robinson and Carville 2010). In particular, the surface water of Jinjang River suffered from leachate pollution (Tiong and Yahya 2005). After almost 15 years of operation, TBL was officially closed in 2006, but the closure did not seem to bring a solution to the leachate contamination and air pollution problem, although restoration projects of the landfill have long begun in 2005. Monitoring of groundwater at the ex-landfill sites at Taman Beringin for identifying emerging contaminants and proper updates of water quality standards and remediation are, therefore, necessary for the management of the environment as well as for the sustainability of water resources in the country of Malaysia. *In this work, emphasis is on groundwater quality and environmental protection. This work therefore focus on identifying the characteristics of the groundwater from previously installed (pre-existing) monitoring wells following standard water quality analysis to provide environmental assessment of the landfill site. Special emphasis is also placed on pollution indicator parameters: COD, BOD, NH_3-N and heavy metals.*

Description of the study area

Taman Beringin ex-landfill site is a 30 hectare (ha) site located at North Jinjang; about 10 km North West of Kuala Lumpur Federal Capital Territory of Malaysia at latitude 03°13.78′ North and longitude 101°39.72′ East. TBL was not designed as sanitary landfill type and was designated municipal solid waste (MSW) landfill by DBKL to accommodate domestic and commercial waste origin collected in the city. The waste level measured at the top of the capped landfill (i.e., plateau area of the landfill) is approximately 52 m in thickness and the ground level in this part of the landfill is about 87.86 mRL (JPSPN and KPKT 2013). The landfill area is mostly surrounded by residential apartments at the very edge and near surroundings. Three of the groundwater monitoring well stations are within the three residential areas. The Taman Beringin solid waste transfer station is located on the north-western flank of the landfill close to monitoring well at BH3 at the north-eastern part, at the bank of the Jinjang River. Figure 1 shows Taman Beringin Landfill site in Kuala Lumpur and the position of monitoring wells in the landfill site.

Taman Beringin landfill site is blessed with a tropical climate. The area gets great levels of precipitation all year round with two monsoons between March–April and October–November coinciding with the southeast and southwest monsoon seasons, respectively. October, November and April are the wettest months of the year and November receives more than 400 mm of precipitation, while March–April gets around 700 mm of rainfall. May and June gets less than 140 mm of rainfall. Total annual precipitation averages 2366.2 mm (93.2 in). The study area also receives a great level of sunshine all through the year;

Fig. 1 Taman Beringin landfill (TBL) site and the position of groundwater monitoring wells

although the temperature varies a little from season to season with the average high of 28–32 °C, while the low hardly falls below mid-20s. In the northern flank of the study area is Jinjang River which is an important distributary of a major upstream, River Batu. River Batu and River Jinjang make up part of the Klang River Basin in Kuala Lumpur. Groundwater modeling in the studied area, confirmed the groundwater flows towards the river/pond in a direction of West to Southeast (see Fig. 1). The aquifer at this site is a shallow sandy type of about 5–10 m thickness and a deeper fractured bedrock limestone. The hydrogeology of the study area is however, characteristic of the Kuala Lumpur Limestone formations which are overlaid by fluvial deposits (Stek 2008). Soil recovered from seven borehole drillings, shows that soil stratum at the top of capped landfill is overlain by rubbish and the strata vary from very stiff sandy silt/clay with depth approximately 8 m thickness to medium dense silty sand at depth of 27.86 mRL with limestone at depth 15.86 mRL (JPSPN and KPKT 2013). The bottom and flat area of the landfill near to the existing transfer station generally consists of sandy silt at approximately 1.5 m thickness at the top stratum. Underlying layers mainly consists of loose to medium dense silty sand underlain by limestone (JPSPN and KPKT 2013).

Materials and methods

Groundwater and leachates sampling

The sampling of the groundwater was carried out during 17–19 December 2012 in some selected groundwater points at the landfill site. Four monitoring wells, previously installed were selected to collect groundwater samples. The choosing monitoring wells have been designated BH3, BH5, BH6 and BH7 and are well positioned and located exactly on the pathway of the groundwater flow in such a way that any contaminants in the groundwater will be captured in all the boreholes (see Fig. 1). The locations of the groundwater points were obtained by a hand held Global Positioning System (GPS) (Table 1). In the first sampling exercise monitoring wells were first being purged by low flow pumping to obtain fresh groundwater for sampling. However, total suspended solid (TSS) and COD readings were inconsistent with the quality of groundwater observed in the field at groundwater points BH3, BH5 and BH6. Therefore, to verify the result obtained for COD and TSS from these groundwater points re-testing and subsequent sampling and analysis were conducted on 11th and 29th January 2013. During the re-testing, the groundwater samples were collected after the extraction of water by

Table 1 Description of the position of the selected groundwater points and water level from Taman Beringin landfill site

Sample code	Distance from landfill[a] (m)	GPS coordinate		Ground level (mRL)	Ground water level (mRL)	Water level (m)
		Latitude	Longitude			
BH3	169.73	N03°13′47.5″	E101°39′42.6″	45.32	43.82	1.5
BH5	341.56	N03°13′37.7″	E101°39′53.2″	46.43	43.23	3.2
BH6	396.68	N03°13′33.6″	E101°39′52.4″	46.77	42.47	4.3
BH7	223.95	N03°13′47.5″	E101°39′42.6″	55.72	45.92	9.8

mRL mean reduced level

[a] Distance—values are with reference from the center of landfill

evacuating the still waters using a foot valve attached to a bailer. The evacuation process removed standing water in the borehole. A minimum of three borehole volumes of water was purged to ensure the samples collected consisted of fresh formation water. Sample collection was carried out as soon as the borehole had recovered to a level sufficient for sampling. Water collected in the sampling bailer was transferred to the sample bottles prepared for the various analyses. Care was taken to minimize agitation and aeration of the water sample during transfer. The respective preservatives were then added to the samples before being kept in shipping cooler and packed with ice to keep the temperature at 4 °C for transportation to the laboratory. Important in situ parameters such as temperature, pH, were tested at the site itself. The necessary hydrogeologic study had consisted of collation and analysis of existing hydrogeologic data on strata logs, groundwater levels; and flow direction. However, to complement this information, depth of groundwater table was measured in each well using a dip meter with respect to the ground surface. Table 1 shows the description of the position of the selected groundwater points and water level from Taman Beringin landfill site.

Composite raw surface leachates from six different points at the base of the landfill were also collected during 17–19 December 2012. The pH, temperature, and sampling of chemical parameters of the leachates were measured at 12-h time interval. The chain-of-custody form was then filled for onward transmission to the receiving laboratories for analysis.

Laboratory and statistical analysis

The analysis of sampled leachates and groundwater samples was carried out by Spectrum Laboratories Sdn. Bhd and E. S. Tech venture Sdn. Bhd. Physico-chemical analyses followed method of APHA (2005). Both the leachate and groundwater samples were analyzed based on their relative importance in terms of field parameters, organic, inorganic and heavy metals components which are characteristic of general constituents in municipal landfill leachate. Physico-chemical parameters examined in groundwater and leachates samples include: color, total dissolved solids (TDS), TSS, total hardness (TH), BOD, COD, cyanide (CN), oil/grease, sulfides (S^{2-}), fluoride (F^-), chloride (Cl^-), NH_3-N, sulfate (SO_4^{2-}) and NO_3^-. Heavy metals include: cadmium (Cd), copper (Cu), iron (Fe), nickel (Ni), lead (Pb) zinc (Zn), manganese (Mn) boron (B), selenium (Se), mercury (Hg) chromium (VI) (Cr^{6+}). Determination of chromium (III) Cr^{3+} was by in house method based on APHA 3120 B, 2005 and APHA 3500-Cr B, 2005 (by calculation). Biological analyses also included total coliform bacteria tests by method of APHA 9221 B, 2005. The data were statistically analyzed by setting up and calculating for the various parameters using Statistical Package for Social Sciences (SPSS 2009).

Results

Results of surface raw leachates analysis

The mean and standard deviation results of some physico-chemical and heavy metal parameters determined in surface raw leachates for six points in Taman Beringin ex-landfill are presented, the minimum and maximum values obtained for these parameters are also listed in Table 2.

The different measurements obtained for the 12-h intervals show no significant variation in pH of the six leachate samples with mean and standard deviation value of 7.8 ± 0.084. The pH of leachate samples from TBL is therefore alkaline. The highest pH value of leachate taken during the sampling was 7.8 and the lowest was 7.6. The mean and standard deviation value (1469.8 ± 619.3) for color (ADMI) in the leachates were much higher than the acceptable conditions for discharge of leachate according to the DOE regulation 2009 (DOE 2010).

The BOD (90.5 mg/L), COD (456.16 mg/L) and TSS (81.5 mg/L), values were relatively high in the leachate samples of Taman Beringin landfill in comparison with DOE regulation 2009. The ratio of BOD/COD of raw leachate samples obtained was 0.19 and remained similar throughout the range of concentrations.

Table 2 Physico-chemical characteristics of surface raw leachates from Taman Beringin landfill and comparison with regulation 2009

Parameters	Unit	Mean ± SD	Minimum	Maximum	Regulation 2009
pH		7.8 ± 0.084	7.6	7.8	6.0–9.0
Color	ADMI	1469.8 ± 619.3	995	2,267	100
COD	mg/L	456.16 ± 102.42	309	586	400
BOD	mg/L	90.5 ± 20.8	60	116	20
BOD/COD		0.19	0.19	0.19	NS
TSS	mg/L	81.5 ± 75.8	21	182	50
Phenol	mg/L	0.008 ± 0.002	0.006	0.011	0.001
Formaldehyde	mg/L	0.10 ± 0.03	0.08	0.14	1.0
Oil/grease	mg/L	10 ± 5.06	4	16	5.0
CN	mg/L	–	ND	ND	0.10
NH_3 -N	mg/L	175.5 ± 73.3	96.2	273	5.0
S^{2-}	mg/L	0.15 ± 0.07	0.1	0.2	0.50
F^-	mg/L	2.72 ± 1.63	0.7	4.4	2.0
Hg	mg/L	–	ND	ND	0.005
As	mg/L	0.08 ± 0.06	0.056	0.152	0.05
Sn	mg/L	–	ND	ND	0.20
Se	mg/L	–	ND	ND	0.02
Zn	mg/L	0.17 ± 0.067	0.081	0.248	2.0
Pb	mg/L	0.07 ± 0.041	0.021	0.123	0.20
Cd	mg/L	0.0018 ± 0.0004	0.001	0.002	0.01
Ni	mg/L	0.031 ± 0.009	0.019	0.043	0.20
Ba	mg/L	0.11 ± 0.03	0.075	0.140	1.0
Fe	mg/L	4.78 ± 0.98	3.327	5.783	5.0
Mn	mg/L	0.27 ± 0.07	0.181	0.355	0.20
Cr^{3+}	mg/L	0.03 ± 0.004	0.02	0.03	0.20
Cr^{6+}	mg/L	–	ND	ND	0.05
Cu	mg/L	0.041 ± 0.019	0.018	0.069	0.20
B	mg/L	0.74 ± 0.35	0.276	1.057	1.0
Ag	mg/L	–	ND	ND	0.10

± denotes standard deviation

Maximum number of samples (n) = 6

Regulation 2009: Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill)

Source DOE (2010)

Contaminants, phenol, oil/grease in raw surface leachates show high concentration of these compounds when compared with the regulation 2009 standard with mean and standard deviation values of 0.008 ± 0.002 and 10 ± 5.06, respectively. The mean value for oil/grease was found to be twice above the compared standard of 5.0 mg/L. Formaldehyde (0.10 ± 0.03) was at low concentrations and did not exceed the standard. The concentrations of CN rarely occurred in raw surface leachates (<0.02 mg/L). Low concentration of S^{2-} was observed in two of the leachates samples, the highest and the lowest being 0.2 and 0.1 mg/L, respectively, with mean and standard deviation values of 0.15 ± 0.07. A high concentration of F^- (2.72 mg/L) which have exceeded the regulation 2009 standard at 2.0 mg/L was observed in the leachate samples. The maximum and minimum values of F^- observed are 4.4 and 0.7 mg/L respectively. Nitrogenous compound such as NH_3-N (175.5 mg/L) was present in higher concentration relative to the compared regulation 2009 standard (5.0 mg/L).

The raw surface leachate samples showed no observable attenuation trend among the heavy metals parameters. Hg, Sn, Se. Cr^{6+}, Ag were generally undetectable, although the mean values obtained for Ba (0.11 mg/L), Fe (4.78 mg/L) and B (0.74 mg/L) are within the stipulated value. However, the maximum values obtained in some of the leachate samples for these heavy metals are slightly above the compared standard. The concentration of Mn (0.27 ± 0.07) and As (0.08 ± 0.06) in raw surface leachates have slightly exceeded the stipulated values of 0.20 and 0.05 mg/L set by the regulation 2009, respectively.

Result of groundwater analysis

Table 1 shows a description of the position of the selected groundwater points and the water level measurement from

Table 3 Physico-chemical and heavy metal characteristics of groundwater samples from Taman Beringin ex-landfill with increased distance from landfill and the comparison with the Malaysian National Drinking Water Quality Standard (NDWQS)

Parameters	Unit	BH3	BH7	BH5	BH6	Mean	NDWQS
Distance from landfill[a]	m	169.730	223.95	341.56	396.68	282.98	
Temperature	°C	29.5	29.0	29.1	29.1	29.1 ± 0.22	NS
pH		7.6	7.7	7.7	7.8	7.7 ± 0.081	6.5–9.0
Color	ADMI	15	149	56	15	58.75 ± 63.19	NS
T.H (EDTA)	mg/L	243	436	354	206	309.75 ± 105.06	500
COD	mg/L	23	196	179	9	101.75 ± 99.42	25(*)
BOD	mg/L	4	37	68	3	28 ± 30.99	3(*)
BOD/COD		0.17	0.18	0.37	0.33	0.26 ± 0.10	NS
TSS	mg/L	–	82	2	–	42 ± 56.56.	50(*)
TDS	mg/L	392	2,200	510	400	875.5 ± 884.63	1,000
NH_3-N	mg/L	12.89	57.30	22.22	32.01	31.10 ± 19.12	0.3(*)
SO_4^{2-}	mg/L	8.4	3.7	195	ND	69.03 ± 109.11	250.0
NO_3^-	mg/L	1.26	2.23	ND	1.92	1.80 ± 0.49	10.0
Cl^-	mg/L	5.4	301	40.8	39.7	96.72 ± 137.17	250
S^{2-}	mg/L	ND	ND	ND	ND	–	NS
F^-	mg/L	0.4	1.0	0.5	1.0	0.72 ± 0.32	0.4–0.6
CN	mg/L	ND	ND	ND	ND	–	0.07
Oil/grease	mg/L	ND	ND	ND	ND	–	NS
Phenol	mg/L	ND	ND	ND	ND	–	0.002
Formaldehyde	mg/L	0.15	0.14	0.17	0.13	0.14 ± 0.017	0.9
Hg	mg/L	ND	ND	ND	ND	–	0.001
As	mg/L	ND	ND	ND	ND	–	0.01
Sn	mg/L	ND	ND	ND	ND	–	NS
Se	mg/L	ND	ND	ND	ND	–	0.01
Zn	mg/L	0.023	0.034	0.487	0.012	0.13 ± 0.23	3.0
Pb	mg/L	0.013	ND	0.094	0.019	0.042 ± 0.045	0.01
Cd	mg/L	ND	ND	ND	ND	–	0.003
Ni	mg/L	0.012	0.018	0.024	0.010	0.016 ± 0.006	0.02
Ba	mg/L	0.048	0.173	0.051	0.041	0.078 ± 0.063	0.7
Fe	mg/L	0.047	0.139	1.442	0.028	0.41 ± 0.68	0.3
Mn	mg/L	0.053	0.082	0.028	0.077	0.06 ± 0.024	0.1
Cr^{3+}	mg/L	ND	ND	ND	ND	–	NS
Cr^{6+}	mg/L	ND	ND	ND	ND	–	NS
Cu	mg/L	0.013	0.044	0.037	0.010	0.026 ± 0.017	1.0
B	mg/L	ND	0.411	0.061	0.004	0.15 ± 0.22	0.5
Ag	mg/L	0.001	ND	ND	ND	0.001	0.05
Coliform	MPN/100 ml	<2	<2	<2	<2	–	NS

± is standard deviation

ND not detected, *NS* not specified, *ADMI* American Dye Manufactures Institute

[a] Distance- value is with reference from the center of landfill; NDWQS- National Drinking Water Quality Standard (NDWQS)

(*) National Water Quality Standards (NWQS) for Malaysia CLASS II A (represent water supply of good quality set for the protection of human health and sensitive aquatic species)

Source: MOH (2004)

Source: EQR (2006)

Taman Beringin ex-landfill site. The summary of analytical results of physico-chemical and heavy metal characteristics of groundwater from Taman Beringin ex-landfill with increased distance from the landfill are presented in Table 3. The comparisons of groundwater quality parameters were made with the National Drinking Water Quality Standard (NDWQS) (MOH 2004). In cases where there are no stipulated values for NDWQS; it was compared with the proposed National Water Quality Standards (NWQS) for Malaysia CLASS II A (EQR 2006).

The result of temperature of the groundwater samples was almost identical with a mean temperature value of 29.1 °C and standard deviation value of ±0.22 which indicates that the temperature is almost close to each other. However, similar temperature measurement (29.1 °C) was recorded for BH5 and BH6. The results of our analysis show a high negative correlation coefficient ($r = -0.7496$) between the depth of water and temperature.

The pH in the groundwater samples range from 7.8 to 7.6. The mean pH value determined in the present study is found to be 7.7 and for the most part values of pH were in close range, with a standard deviation of ±0.081. However, alkaline pH is observed to be maintained in all the wells but the pH was more stable at 7.7 in BH 5 and BH7. The pH values and mean pH recorded for the groundwater in this study are within the stipulated pH tolerance range of 6.5–9.0 set by the Malaysian National Drinking Water quality standard (NDWQS).

The levels of TSS in groundwater samples from Taman Beringin are relatively at low concentrations and completely undetectable in some of the wells, with the exception of the high value recorded in BH7 (82 mg/L). The mean TSS value of groundwater recorded is 42 mg/L and within the stipulated CLASS II A standard. However, among the individual wells groundwater from BH7 has exceeded the TSS stipulated range. Attenuation of TDS concentrations could be observed in down gradient wells between BH7 through BH6. On the other hand, a relatively high TDS value of 2,200 mg/L was recorded in BH7 and decreases sharply in the other monitoring wells. The trend in TDS recorded in the groundwater samples between BH5 and BH6 decreases within similar range of 100 mg/L. The average value of TDS determined in the groundwater samples from Taman Beringin is 875.5 mg/L with a standard deviation value of ±884.63. The mean value of TDS is within the compared standard except for groundwater samples from BH7 which is more than twice the NDWQS value of 1,000 mg/L. The measured TH of the groundwater samples was found to be decreasing at a similar concentration of 105 mg/L with increased distance downgradient the landfill. The exception is upstream well at BH3 (243 mg/L) which is relatively higher in concentration compared to downgradient well at BH6 (206 mg/L). Therefore, with a mean TH value of 309.75 mg/L the groundwater beneath the Taman Beringin landfill is said to be hard. However, the mean value is below the NDWQS stipulated value of 500 mg/L

BOD values recorded in each of the individual well sampled as well as the mean BOD value (28 mg/L) have exceeded the NWQS (Class II A). The exception is BH6 which is at the same concentration of 3 mg/L with the level stipulated by the standard. The trend in the level of COD assumes an almost similar pattern of BOD decreasing from upstream to downgradient wells, but at much reduced levels at the furthest downgradient well at BH6, although, there is no definite pattern observed. However, the concentrations observed in COD levels were highest in BH7 (196 mg/L) and BH5 (179 mg/L). It was observed that the values recorded for COD in all the wells sampled were relatively higher than the corresponding BOD values of the same well. The mean and standard deviation of COD were found to be 101.75 ± 99.42 and these values are by far above the NWQS (CLASS II A) stipulated value. The

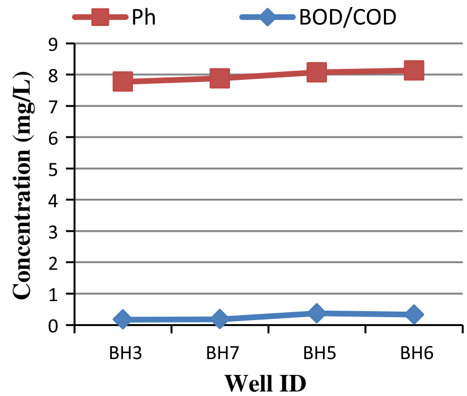

Fig. 2 Changes in pH with BOD/COD ratio of groundwater samples from Taman Beringin ex-landfill sites

mean BOD/COD value determined in groundwater samples is 0.26 which signify generally low organic content of the groundwater samples. However, the general pattern observed in the BOD/COD ratios of the groundwater samples indicates a slow biodegradability throughout the landfill. Coefficient value of data set between BOD/COD and temperature indicates moderately correlated relationship ($r = 0.6377$) indicating that pH increases (although slightly) with increasing biodegradability and vice versa. Figure 2 shows the changes in pH with BOD/COD ratio of groundwater samples from Taman Beringin ex-landfill site.

Oil/grease and organics, such as CN and phenol were rarely detectable with the exception of Formaldehyde. The limit of detection (LOD) of parameters CN and Phenol are 0.02 and 0.001, respectively. The mean concentration recorded for organic compound Formaldehyde is 0.14 mg/L and the values in all the wells are close with a standard deviation of ±0.017. The values are well within the NDWQS.

The measured values of SO_4, and NO_3, in groundwater were below the NDWQS stipulated value for each of the anion for potable water (Table 3). The exceptions in this group of anions are NH_3-N, Cl^- and F^-. NH_3-N was recorded in relatively high concentrations in all of the wells sampled. The highest value recorded is 57.30 mg/L in BH7 and the lowest is 12.89 mg/L in upstream BH3. The level in BH6 (32.01 mg/L) about 396.68 m away from landfill is higher relative to BH5 (22.22 mg/L) which is closer to the landfill at a distance of 341.56 m. However, there is evidence of biological oxidation process of NH_3-N (nitrification) which could be related to the presence of NO_3 in the groundwater aquifer (the exception is the aquifer in BH5). The concentration of NO_3 in groundwater samples were relatively at low level with mean and standard deviation value at 1.80 ± 0.49. In contrast, the NH_3-N concentration of all the groundwater samples from TBL and the mean concentration (31.10 mg/L) is far above the stipulated

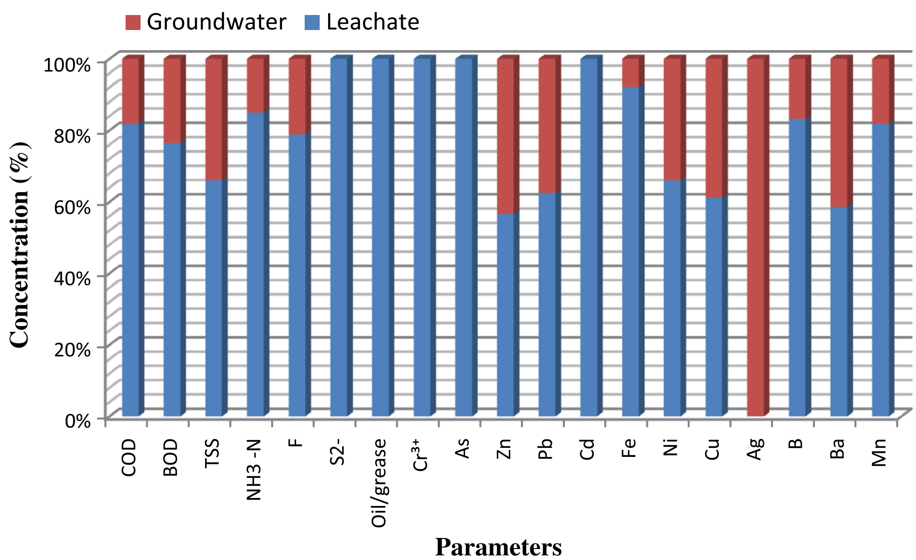

Fig. 3 Comparison of the mean concentration (%) of some chemical and heavy metal parameters of the raw surface leachate with groundwater sample from Taman Beringin landfill site

value of 0.3 mg/L of the NWQS (ClASS II A). The concentration of F^- (1 mg/L) recorded in BH6 and BH7 was similar and the value obtained in these two wells and their overall mean concentration value (0.72 mg/L) obtained are above the range set by NDWQS (0.4–0.6). Exceptionally high Cl^- concentration (301 mg/L) was recorded for groundwater samples of the aquifer around BH7. This well (BH7) is the nearest of the downgradient well with increased distance from the center of the landfill. However, there is an abrupt washout pattern of Cl^- concentration falling from 301 mg/L in BH7 then continuing at few concentrations of about 40.8 mg/L in BH5 and onwards were it appears fairly stable at 39.7 mg/L in BH6. The permissible Cl^- concentration allowed in groundwater according to NDWQS is 250.0 mg/L but the Cl^- concentration of the groundwater sample around BH7 has exceeded the stipulated Cl^- level permitted.

In the case of trace metals in the groundwater samples from TBL, Hg, As, Sn, Se, Cd, Cr^{3+}, Cr^{6+}, Ag rarely occurred at concentrations large enough to be detected. The limits of detection (LOD) of these heavy metal parameters are: Hg (<0.001), As (<0.01), Sn (<0.01), Se (<0.01), Cd (<0.001), Cr^{3+} (<0.02), Cr^{6+} (<0.02), Ag (<0.001). In particular, Fe (0.41 mg/L); Pb (0.042 mg/L and Ni (0.016 mg/L) occurred at concentrations above the compared standard. Exceptionally high content of Fe (1.442 mg/L) was observed in BH5, while Pb (0.094 mg/L) and Ni (0.024 mg/L) were equally highest in the same well. The acceptable level of 0.3, 0.01, 0.02 mg/L for Fe, Pb and Ni, respectively, set by NDWQS in Malaysia was not met in some of the wells. Mn, Cu, B, Zn, Ba, on the other hand, occur in trace concentration. Figure 3 shows a comparison of the mean concentration (%) of some chemical and heavy metal parameters of the raw surface leachates and groundwater samples from Taman Beringin landfill site.

Discussion

The high negative correlation coefficient $r = -0.7496$ of the relationship of temperature and water levels is due to the fact that groundwater around BH3 is warmest due to the shallow water table of 1.5 m which is sufficiently small for the temperatures within depths to cause slight increase in the warming of the water. BH6 has a temperature similar to BH5 (29.1) due to thicker water table but the shallowest depth compared to BH7 (refer to Table 1).

The general alkaline pH of 7.8 of raw surface leachate obtained in this study is indicative of landfill due to methanogenic reactions and that solubilisation of the majority of organic components is near completion and approaching stabilization stage. The result of pH obtained in this study is also in good agreement with the results obtained for raw leachate in TBL by Agamuthu (2001) which was in the high range of 8.07–8.50. However, these two results (Agamuthu 2001 and the present study) show that the pH of raw leachates has decreased. Furthermore, the two results further signify the evolution of the methanogenic phase of the landfill in a space of 13 years. Although the pH remains relatively within a close range throughout the sampling points in both groundwater and leachates samples from TBL, however, the relative high value of 7.8 observed in the groundwater sample in BH6 can be attributed to the changes in concentration of some parameters within the landfill. Overall the general alkaline pH of the groundwater samples (7.7) which is higher than natural groundwater pH reflects the influence of the leachate contaminants on the groundwater.

The highest color value obtained in BH7 (149 ADMI) corroborates with the highest TSS of 82 mg/L of the groundwater samples in the same well indicating that suspended materials have contributed to the changes in the apparent color of the groundwater. Moreover, this high color value also signifies that BH7 is the most contaminated groundwater aquifer in this study. Groundwater with high color and TSS as those around BH7 is not suitable for domestic use and this should be avoided. Therefore, groundwater with high color and TSS value around BH7 needs to undergo treatment before it can be used. On the other hand, TDS is a good indicator of the mineralized character of the water especially when in contact with leachate. TDS comprises inorganic salts such as Cl and SO_4. The relatively higher value of TDS (2,200 mg/L) in BH7 may not be unconnected with the high value of Cl in the same well at 301 mg/L. In similar vein, SO_4 concentration of 195 mg/L has contributed to the high TDS value of 510 mg/L in BH5. High TDS value (2,200 mg/L) and TSS (82 mg/L) obtained in the present study in groundwater sample in BH7, which have exceeded the NDWQS of 1,000 and 50 mg/L, respectively, is not considered good water for domestic use or consumption and the groundwater quality represents a significant threat to public health and a potential source of cross-contamination of pollutants to other wells and surface waters, especially rivers around the landfill site. Due to this concern, groundwater drawn from BH7 well should be avoided.

The range of BOD (560–1,520 mg/L) and COD (2,050–5,230 mg/L) of raw leachates recorded by Agamuthu (2001) in TBL where much higher compared to the range of BOD (60–116 mg/L) and COD (309–586 mg/L) of raw leachates in this present study. These two data give a good indication of the strength and characteristics of the leachates which could be attributed to the age of the Landfill then in 2001 and now in 2013. Leachates with high BOD levels above the stipulated standard for discharge could cause a ripple effect on the receiving water and the oxygen may diminish to levels that are detrimental to most aquatic animals.

The measurable COD values recorded in all the groundwater samples could be attributed to result of oxygen demand of microbial oxidation particularly of the NH_3-N oxidation to NO_3 with the exception of BH5. However, the relatively high COD values in BH7 (196 mg/L) and BH5 (179 mg/L) compared to the other wells in the landfill site is connected to anaerobic-reducing condition caused by the COD exerted by ammonia oxidation in the groundwater samples in BH7 and BH5 and the COD that may have been exerted by oxidizable metals such as Fe and Mn. The BOD/COD ratio of raw leachates (0.19 mg/L) that was obtained is indicative of the organic strength of leachates at the landfill site that were analyzed. Thus, the leachates are considered less biodegradable and of intermediate type, produced during the methanogenic phase of the landfill. However, the results of BOD/COD of raw leachates were similar throughout the whole concentrations. This is considered good evidence of sustained biodegradability. In contrast, the ratio of BOD/COD of the groundwater wells (0.26) indicates low biodegradability, and is erratic and unsustained throughout the groundwater wells. This is due to the anaerobic condition prevailing around BH5 through BH6. The recorded BOD/COD values of 0.37 and 0.33 in BH5 and BH6, respectively, show that there are still organic materials in the groundwater which are still biodegradable but likely, some condition in the wastewater may be inhibiting bacteria that use organic materials. The result of biodegradability of the ratios of BOD/COD obtained in BH3 (0.17) and BH7 (0.18), respectively, shows that the organic materials have reached near stabilization and are unaffected by the condition prevailing around BH5. The Correlation coefficient values ($r = 0.6377$) between BOD/COD and temperature which show moderately correlated with pH signify that the condition of biodegradability in the landfill cannot be attributed solely to temperature condition. However anaerobic-reducing condition that was observed in the landfill around BH5 plays a major role in the decomposition process in the groundwater aquifer. The point of an anoxic condition prevalent of the groundwater samples around the aquifer in BH5 is further strengthened by the undetectable NO_3 (<0.02 mg/L) in the groundwater samples. In addition, the relatively high Fe content (1.442 mg/L) in the same well at BH5 may be the result of reduction of Fe^{3+} to Fe^{2+} and this is also an additional confirmation to the anaerobic-reducing condition in that aquifer. According to Appelo and Postma (2005) high concentrations of redox sensitive species such as Fe and Mn can as such be attributed to reducing conditions. However, more testing is in progress to confirm these conditions within the Taman Beringin landfill. Further investigation is also needed to determine the level of some aquifer parameters such as dissolved oxygen (DO) and oxygen-reducing potential (ORP) and this will give a good representation of the condition of the groundwater aquifer.

The level of NH_3-N in groundwater samples from 57.30 to 12.89 mg/L are lower compared to the mean value of raw leachates (175.5 ± 73.3). This may be attributed to the retard action of the water bearing media on NH_3-N concentrations (i.e., NH_3-N adsorption) as they migrate through the aquifer to the groundwater. According to Robinson and Carville (2010), during the active phase of the landfill at Taman Beringin, in the early years of waste treatment, leachate was strong and methanogenic, and contained in excess of 1,000 mg/l of NH_3-N. This difference in the leachate concentrations compared to the levels in this study is purely due to the age of the landfill. Although, the levels of NH_3-N in raw leachates in this study are much lower compared to other studies mentioned above, yet the concentrations have exceeded the acceptable limit for raw leachate set at 5.0 mg/L by DOE regulation 2009. Furthermore, the levels of NH_3-N recorded in all the groundwater wells were also observed to be above the compared NWQS (CLASS II A) standard of 0.3 mg/L. Groundwater and raw leachates in Taman Beringin will require routine treatment before it is discharged or used because high concentrations of NH_3-N could be a substantial source of cross-contamination of nitrogen related compounds in receiving water. However, the prevalent concern of NH_3-N of their health effect in groundwater is much related to the presence of Cl^-. High level of Cl^- is recorded at the groundwater at BH7 at 301 mg/L which could equally increase the toxicity of NH_3-N. Cl^- and NH_3-N could form ammonium chloride and chloramines which is also toxic at concentration above the required level. Furthermore, NH_3-N attenuation products such as nitrate (NO_3) and nitrite (NO_2) are also of concern of the health risk for infant regarding methaemoglobinaemia, or so called "blue-baby syndrome". In TBL, downgradient wells show decrease in NH_3-N concentrations (although only over a few hundred meters away from the landfill) but it suggest that attenuation of NH_3-N is occurring along the flow path with the exception of BH5 where nitrification process is absent due to the inhibiting anoxic condition of the groundwater. Evidently, it appears that biological oxidation is not the important process in BH5 but a chemical-reducing condition.

The non detectable heavy metals Hg, Sn, Se, in raw surface leachate from Taman Beringin landfill corroborate with their absence in groundwater and is an indication that the Taman Beringin landfill did not receive waste containing these metals. The influence of high pH (e.g., alkaline) cannot be ruled out and may have exerted direct influence on the low concentration of some of the heavy metals such as Pb, Cd, Mn, Ni, Cr^{3+} with the exception of Fe which are exceptionally higher in both raw leachates and groundwater samples. At alkaline pH water can hold fewer of these metals in solution because heavy metals in groundwater are most soluble at low pH. Even though there may be migration of contaminants into groundwater, as observed for Zn Pb, Cd, Cu, Mn, Ni and B in Taman Beringin the levels were low with the exception of Fe in groundwater and leachate at 1.442 and 5.783 mg/L, respectively. The value was considered far higher in raw drinking water above the 0.3 mg/L as stipulated by NDWQS, while the maximum value in leachate samples was slightly above the DOE regulation 2009 standard of 5.0 mg/L for the discharge of raw surface leachate. Higher concentration of Fe is not uncommon in Taman Beringin landfill leachate as these also agree with the value recorded by Agathamu (2001) which was in the range of 7–9 mg/L. High Fe in water supply is well known for causing reddish-brown stainings that causes corrosion. Therefore, the water will need to undergo routine chemical treatment before it can be applied for domestic use or drinking purpose. Concentration of Pb (0.094 mg/L) and Ni (0.024 mg/L) in BH5 at much higher level than the well in BH3, BH7 and BH6 and above the stipulated standard for raw drinking water. This may be due to the reducing condition in BH5 which favors the mobility of the above mentioned metals. Overall, the metals As, Pb, Mn, Ni, and Fe are the heavy metals that have occurred in concentration higher than the permissive level according to NDWQS and DOE regulation 2009 for raw drinking water and discharge of leachate, respectively. These heavy metals (excluding Ag) were identified in the leachate samples and at the same time have spread to the groundwater wells (see Fig. 3) indicating the impact of leachates contamination on groundwater quality beneath the landfill.

Recommendation

The groundwater quality of the study area can be improved by:

1. Preventing the existing leachate from leaking into the river via groundwater flow. This could be done by installing pumping wells along the river to extract the contaminated groundwater at downgradient of the landfill. Additional monitoring wells should be installed in the upstream to monitor background water quality. However, a cut-off wall may also be suggested but may not be practical at this time.
2. Reducing the leachate generation: Investigating the efficiency of the influence of the sub-soil stratigraphy and hydrogeology condition for containing leachate outflow in the study area which is vital for groundwater protection could be a confirmatory step for promoting natural attenuation especially for ammonia–nitrogen and also further decreasing the COD and BOD level in the groundwater.
3. Improving the quality of the leachate: From the mentioned mean results of BOD/COD ratio of raw leachates (0.19) and groundwater samples (0.26), the leachate will be easy to treat biologically but in light of the presence of other contaminants such as the heavy metals it is necessary to undergo a combined process of routine chemical treatment before the routine biological treatment. Leachates with BOD/COD ratio of 0.19 as observed in TBL provide a good effectiveness of chemical treatment, not only for color and metal removals but also for COD reduction.

Conclusion

The groundwater aquifers beneath the Taman Beringin landfill are contaminated by organics and heavy metals and there is a great tendency for an increase in groundwater contamination around the landfill location considering the current levels of contaminants: COD, BOD, TSS, TDS, NH_3-N, Cl^-, F^-, Pb, Ni, and Fe found in the groundwater which have exceeded the stipulated NDWQS in Malaysia. As much as these contaminants are found in the groundwater at concentrations higher than the standard the risk exists especially when the groundwater flows into river/ponds and pollute drinking water and other groundwater resources. Evidently, landfill leachates wastewater is a potential point source responsible for degraded river water in Malaysia. It is therefore recommended that remediation should be done in a responsible way to prevent further spreading of leachates especially into rivers via groundwater flow by reducing the leachate generation and improving the leachate quality to minimize the effects on the surrounding environment.

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