

ASSESSMENT OF VARIOUS TROPICAL MUNICIPAL LANDFILL LEACHATE CHARACTERISTICS AND TREATMENT OPPORTUNITIES

AZIZ S.Q.^{1,*} AZIZ H.A.² BASHIR M.J.K.³ MOJIRI A.²

¹Department of Civil Engineering, College of Engineering Salahaddin University—Erbil, Iraq ²School of Civil Engineering, Engineering Campus USM, 14300 Nibong Tebal, Penang, Malaysia ³Faculty of Engineering and Green Technology (FEGT), Universiti Tunku Abdul Rahman 31900 Kampar, Perak, Malaysia

Received: 01/10/2014 Accepted: 27/05/2015 Available online: 29/05/2015

*to whom all correspondence should be addressed:

e-mail: shoker71@yahoo.com

ABSTRACT

Raw municipal landfill leachate is extremely polluted wastewater and it regards as one of the drawbacks of the sanitary landfill treatment method. If the untreated landfill leachate is discharged to the natural environment, a great problem for the environment can be created, particularly for the water resources. To assess fresh leachate; collection, analyzing for various parameters, and comparing with the standards are essential. Thus, this study was purposed to examine the characteristics of different landfill leachate samples collected from three tropical landfill sites. The results of the formed leachate at the anaerobic Kulim Sanitary Landfill, semi-aerobic Pulau Burung Landfill Site, and anaerobic Kuala Sepetang landfill leachate in the northern region of Malaysia have been analyzed for 27 parameters and compared. The studied parameters in the present study were phenols, zeta potential, oxidation-reduction potential (ORP), chemical oxygen demand (COD), biochemical oxygen demand (BOD₅), heavy metals, nitrogen compounds, salinity, electrical conductivity etc. For checking the risks of the leachate on the environment, the obtained results were compared with the Malaysia Standards. In addition, the leachate treatment opportunities upon the characterization are highlighted in this study. The effectiveness of various applications in treating leachate collected from municipal landfill was presented and discussed. It could be concluded that the knowledge of leachate quality is particularly significant in choosing an appropriate treatment techniques

Keywords: MSW, Landfill leachate, Treatment techniques, anaerobic, semi-aerobic, environment

1. Introduction

Municipal solid waste (MSW) disposal methods include open dump, sanitary landfill, incineration, composting, grinding, etc. Sanitary landfill is the most popular MSW disposal method due to such benefits as easy disposal procedure, low cost, and landscape-restoring effect on holes from mineral workings (Davis and Cornwell, 2008). However, the production of highly contaminated leachate is a major drawback of this method (Aziz et al., 2010, Mojiri et al., 2013).

Landfill leachate is a highly contaminated liquid produced by the percolation of precipitation water through an open landfill or through the cap of a completed site. Leachates may contain large quantities of organic contaminants, ammonia, suspended solid, heavy metals, and inorganic salts, phenols and

phosphorus. If not treated and carefully disposed, leachate could be a major source of water contamination (Aziz et al., 2010; Renou et al., 2008; Uygur and Kargi, 2004; Kiayee, 2013).

El-Fadel *et al.* (2002) recognized that the characteristics of landfill leachate can show substantial spatial and temporal variations depending upon the site, management practices, refuse characteristics (i.e. age and composition), and internal landfill processes. Moreover, the variation of leachate characteristics in MSW can be attributed to several interacting reasons such as the composition and depth of MSW; decomposition and age of MSW; degree of compaction; landfill design and operation; liner (top and base) design and operation; MSW filling procedures; the obtainability of moisture content; accessible oxygen; rate of water movement and temperature (Qasim and Chiang, 1994). Subsequently, leachate characteristics may change from time to time and site to site owing to the parameters such as moisture content, waste composition, temperature, climatic changes etc. (Rafizul and Alamgir, 2012).

The present study was aimed to characterize the leachate from various landfill sites in the northern part of Malaysia i.e. anaerobic Kulim sanitary landfill (KLS), semi-aerobic PulauBurung sanitary landfill (PBLS) and the anaerobic Kuala Sepetang landfill site (KSLS), and hence compared with those results of similar cases available in the literature. The risks of the landfill leachate on the natural environment are determined by comparing leachate quality with the standards of Malaysia. The characteristics results are essentially required in order to propose an adequate treatment method for the studied landfill leachate that considers all the measured pollutants. Furthermore, the performances of various treatment technologies conducted by the authors in recent years were summarized and discussed.

Currently, there are over 230 landfill sites in Malaysia, mostly old dumpsites. Most are simply dumping grounds without any environmental protection. The resulting leachate is discharged directly into water courses without any treatment, which can threaten the surrounding ecosystem, particularly in cases where landfills are located upstream of water intakes.

2. Materials and Methods

2.1. Landfill sites location and characteristics

The current study was performed on the leachate collected from three solid waste landfill sites located at the Northern part of Malaysia. Site location and characteristics of each site are presented as following.

2.1.1. Kulim landfill Site (KLS)

KLS is located in the town of Kulim, Kedah, in the northern area of Malaysia, with geographical coordinates of 5° 25′ 0″ N and 100° 37′ 0″ E. It is surrounded by a palm oil plantation. KLS has a total area of 56 ha, receives approximately 240 tons of municipal solid wastes daily, with a depth of about 20 m. This controlled open dumping site began operation in 1996. KLS is classified as an anaerobic landfill. Yamamoto (2002) and Matsufuji et al. (1993) reported that in an anaerobic landfill, solid wastes are dumped in an excavated area of a plane field, which is filled with water in an anaerobic condition. Typically, anaerobic sanitary landfills are recognized by its sandwich- shaped cover. The produced leachate is conveyed by a corrugated HDPE pipe with a diameter of 30 cm to a lined collection pond, where it remains for specific retention time. Subsequently, it is disposed to the natural environment without any treatment. Leachate produced from this landfill was characterized as having low biodegradability ratio and high concentrations of COD and NH₃-N.

2.1.2. Pulau Burung Landfill Site (PBLS)

PBLS is situated within the Byram forest reserve at 5º 12' 12.1" N latitude and 100º 25' 30.2" E longitude in Penang, about 20 km southeast of Penang Island, Malaysia. The total area of the landfill is 63.7 ha and it is receive about 2,200 tons of solid waste per day. The site was developed in 1991 as a semi-aerobic sanitary landfill level II by establishing a controlled tipping technique. In 2001, it was upgraded to a sanitary landfill level III by using controlled tipping with leachate recirculation. This site has a normal

marine clay liner. It was developed semi-aerobically and is one of the only three sites of its kind in Malaysia. Typically, semi-aerobic landfills have a leachate collection duct. The opening of the duct is surrounded by air, and the duct is covered with small crushed stones. Moisture content in solid waste is small, and oxygen is supplied to the solid waste from the leachate collection duct (Bashir *et al.*, 2009; Yamamoto, 2002; Matsufuji *et al.*, 1993). PBLS generates a dark black-green liquid, which could be categorized as stabilized leachate with high concentrations of COD and NH₃-N and low BOD₅/COD ratio.

2.1.3. Kuala Sepetang Landfill Site (KSLS)

KSLS is located at 4° 49′ 20.08″ N and 100° 40′ 44.08″ E near the town of Taiping, Perak state, Malaysia. The total area of the landfill is 12 ha and it is equipped with a leachate collection pond. In 2007, the landfill received about 300 tons of solid waste daily. Kuala Sepetang landfill is classified as an improved anaerobic landfill. The landfill is more than 16 years old. Some recycling is practiced at the site, mainly by scavengers, and then the remaining waste is dumped in individual phases. Local material is used as cover for dumped waste. The leachate is collected in pools which act as detention ponds. However, no further treatment is done (Aziz *et al.*, 2010). Low BOD₅/ COD and high concentrations of COD and NH₃-N were the characteristics of the leachate in this landfill.

2.2. Leachate sampling and characteristics

Leachate samples were collected from KLS, PBLS, and KSLS landfills in the northern region of Malaysia. The raw leachate samples were taken from collection ponds at KLS, PBLS, and KSLS.

The samples were immediately transported to the laboratory, and were stored in a cold room at 4 °C prior to use for experimental purposes to minimize biological and chemical reactions. The collected samples were characterized as following:

The 4-aminoantipyrine method (8047-Hach) was used to measure phenols by determining all ortho- and meta-substituted phenols or napthols. Zeta potential (mV) was measured using The Malvern Zetasizer Nano ZS. Oxidation reduction potential (ORP) (mV), pH, electrical conductivity (EC) (ms cm⁻¹), temperature (°C), and salinity (g l⁻¹) were measured using multi-paramete analyser. Color (Pt.Co) was measured at 455 nm wavelength using Hach color method 8025. COD (mg l⁻¹) concentration was measured using Hach DR 2800 Spectrophotometer at 620 nm wavelength. BOD₅ (mg l⁻¹) was determined using 5-Day BOD Test (5210-B). Ammoniacal nitrogen (mg I⁻¹ NH₃-N Ness) was determined using Nesslerization method (4500-NH₃) while TSS was determined by using Method No. 2540D. Suspended solids were measured using spectrophotometer with program 630. Acidity (mg l-1 CaCO₃), total alkalinity (mg l⁻¹ CaCO₃) were measured using titration method. Other parameters such as Nitrate –nitrogen (NO₃-N) (mg l⁻¹ NO₃ -N), NO₂-N (mg l⁻¹ NO₂-N-HR), Total phosphorus (mg l⁻¹ PO₄³⁻ -TNT), Magnesium (mg l⁻¹ CaCO₃), calcium (mg I¹ CaCO₃), chloride (mg I⁻¹ Cl⁻), sulfide (mg I⁻¹ S²⁻), Total iron (mg I⁻¹ Fe), and Zinc (mg l⁻¹ Zn) were measured via Hach DR2800 spectrophotometer. The parameter measurements were conducted according to the Standard Method of Water and Wastewater (APHA, 2005). To identify the environmental risks of leachate, the obtained parameter values were compared with the Malaysian standard as per the "Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, under the Laws of Malaysia-Malaysia Environmental Quality Act 1974"(Environmental Quality, 2009).

2.3. Leachate treatability

Based on the characteristics of leachate and the literature review concerning leachate treatment various process can be used to treat landfill leachate. Consequently, the main aim of this section was to evaluate the performance of various processes (e.g., biological, adsorption, advanced oxidation processes, ion exchange, and coagulation flocculation) carried out in recent years to treat landfill leachate generated from a single landfill site in Malaysia (e.g., Pulau Burung Landfill Site).

3. Results and discussions

3.1. Leachate characterization

The characteristics of landfill leachates at KLS, PBLS, and KSLS are given in Table 1. To identify the environmental risks of the leachates, the measured pollutants values values were compared against Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 [Act 127].

3.1.1. Kulim landfill leachate

Kulim landfill leachate contains high concentration of COD (1295 mg l^{-1}) and color (3029 Pt.Co) due to the presence of high molecular weight organic compounds. The concentration of NH₃-N was also high (562 mg l^{-1}). Average BOD₅ of 285 mg l^{-1} was recorded, which gave a low biodegradability ratio (BOD₅/COD) of only 0.201. Moreover, the concentration of phenols, color, suspended solids, BOD₅, COD, NH₃-N, and sulfide all surpassed the allowable limits set in Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 [Act 127]. However, pH, temperature, total iron, and zinc were within the allowable limit. Wide range of some measured parameters, for instance color, returned to the effect of rainfall which caused dilution of generated landfill leachate at the collection pond. In conclusion, Kulim landfill leachate considered as low refractory compounds (BOD₅/COD=0.201) and high concentrations of COD and NH₃-N.

Kurata et al., (2008) measured the concentrations of 41 phenols in landfill leachates from 38 MSW landfill sites in Japan. The main phenols detected by the researchers were phenol, 4-tert-butylphenol, 4tertoctylphenol, 4-nonylphenol, three cresols, bisphenol A, and some chlorophenols. They stated that the sources of phenol and p-cresol were considered to be incineration residues, and the main origin of bisphenol A, 4-tert-butylphenol, and 2, 4, 6-trichlorophenol was considered to be solidified fly ash. On the other hand, the main origins of 4-nonylphenol and 4-tert-octylphenol were considered to be incombustibles. The discharge of leachates to the natural environment without adequate treatment can cause environmental pollution due to high concentration of phenols. Particularly, the disposal of incineration residues including solidified fly ash and the co-disposal of incombustibles and solidified fly ash can increase the opportunity of environmental pollution. A series of traditional wastewater treatment processes can remove phenol, 4- nonylphenol, and bisphenol to an acceptable level (Kurata et al., 2008). Because of its negative effects on public health and ecosystems, phenol and its related compounds have been given high priority in governmental regulations in many countries (Ahn et al., 2008; Kurata et al., 2008; Kujawski et al., 2004; Tziotzios et al., 2007; Xiao et al., 2006). In this research, 4-aminoantipyrine method was used to measure phenols by determining all ortho-substituted and meta-substituted phenols or napthols, but not para-substituted phenols. If not treated and disposed safely, landfill leachate could be a major source of water contamination because it could percolate through soil and subsoil, causing high pollution to receiving waters (Aziz et al., 2010). Thus, treatment of risky leachate constituents before discharge has been made a legal requirement to prevent pollution of water resources and avoid both acute and chronic toxicities. Umar et al. (2010) had reported characteristics (pH = 7.8, BOD₅ = 515 mg l^{-1} , COD =1593 mg l^{-1} , BOD₅/COD = 0.32, NH₃-N = 503 mg l^{-1} , total iron = 6 mg l⁻¹ and zinc = 0.3 mg l⁻¹) of Kulim landfill leachate. The obtained results (particularly the parameters pH, BOD₅, COD, BOD₅/COD, NH₃-N, total iron, and zinc) generally were in agreement with the published data by Umar et al., (2010).

Table 1. Characteristics of raw landfill leachate at KLS (11 samples), PBLS (10 samples), and KSL (4 samples)

		Landfill leachate at										
No.	Parameter	Kulim site (Values)			Pulau B	Surung site (\	/alues)	Kuala Se	Discharge			
		Minimum	Maximum	Average	Minimum	Maximum	Average	Minimum	Maximum	Average	Limit ^a	
1	Phenols (mg l ⁻¹)	0.14	5.25	1.54	2.85	10.50	6.95	0.96	2.14	1.56	0.001	
2	Zeta potential (mV)	-16.90	-13.40	-14.87	-20.40	-19.70	-20.05	-21.80	-19.90	-20.85		
3	ORP (mV)	5.70	13.60	11.25	-220.50	-135.00	-177.75	-230.00	-80.00	-161.00		
4	рН	6.93	8.81	8.02	8.14	8.92	8.36	8.21	9.48	8.66	6 to 9	
5	Color (Pt.Co)	700	7475	3029	2310	4790	3615	3040	9090	6398	100	
6	EC (ms cm ⁻¹)	4.45	13.92	7.66	21.20	23.57	22.36	6.34	11.94	9.68		
7	Temperature (°C)	31.00	37.00	33.60	31.44	37.00	34.66	29.33	36.50	33.46	40	
8	Total solids (mg I ⁻¹)	1552	10568	4832	8660	11084	9507	5236	7248	6615		
9	Suspended solids (mg l ⁻¹)	156	1374	553	374	1372	815	512	960	693	50	
10	Acidity (mg l ⁻¹ CaCO ₃)	600	4800	1984	1960	7600	3544	700	1100	900		
11	Total Alkalinity(mg I ⁻¹ CaCO ₃)	8000	22000	14667	23600	64400	42342	11000	23400	16000		
12	Total Hardness (mg l-1 CaCO ₃)	620	2780	1770	840	3460	1559	560	1270	823		
13	BOD ₅ (mg l ⁻¹)	47	1024	285	71	336	181	203	349	257	20	
14	COD (mg l ⁻¹)	290	2860	1295	1580	2030	1819	752	1850	1456	400	
15	BOD ₅ /COD	0.09	0.40	0.201	0.04	0.19	0.100	0.12	0.28	0.19	0.05	

Table 1. Characteristics of raw landfill leachate at KLS (11 samples), PBLS (10 samples), and KSL (4 samples) (Cont.)

		Landfill leachate at									Standard
No.	Parameter	Kulim site (Values)			Pulau Burung site (Values)			Kuala Sepetang site (Values)			Discharge
		Minimum	Maximum	Average	Minimum	Maximum	Average	Minimum	Maximum	Average	Limit ^a
16	NH ₃ -N (mg l ⁻¹ NH ₃ -TN)	360	1039	562	1145	2150	1627	455	700	564	5
17	NO ₃ -N (mg I ⁻¹ NO ₃ –TN)	2.5	30	13.38	9	27	20.33	3.5	15	9.83	
18	NO ₂ -N (mg I ⁻¹ NO ₂ -HR)	5	110	53.9	4	140	59.67	20	100	55.33	
19	Total phosphorus (mg l ⁻¹ PO4 ⁻³ TNT)	8	140.8	32.25	10	56.6	28.58	24.4	61.8	44.63	
20	Magnesium (mg l ⁻¹ Mg-CaCO ₃)	90	480	271.11	230	800	410	80	360	203.33	
21	Calcium (mg l ⁻¹ Ca-CaCO ₃)	440	2460	1498.89	40	3180	1148.89	390	910	620	
22	Chloride (mg l ⁻¹ Cl ⁻¹)	160	425	313.44	390	2237	994.11	397	1710	899.33	
23	Sulfide (mg l ⁻¹ S ²⁻)	0.1	1.92	0.68	0.2	0.48	0.35	1.07	1.31	1.19	0.5
24	Total iron (mg l ⁻¹ Fe)	0.5	11.4	3.82	0.9	16.3	4.9	2.2	4.9	3.43	5
25	Zinc (mg l ⁻¹ Zn)	0	1.15	0.33	0	2	0.52	0.15	0.3	0.22	2
26	Salinity (g l ⁻¹)	2.35	4.5	3.43	12.7	14.11	13.62	3.5	6.72	5.45	
27	TDS (%)	2.9	5.23	4.17	15.18	15.34	15.26	4.28	7.74	6.17	

^a Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 [Act 127]⁻

3.1.2. Pulau Burung landfill leachate

Pulau Burung landfill leachate had high-intensity color (3615 Pt.Co) and high COD concentration (1819 mg l⁻¹) (Table 1). Average BOD₅ value of leachate from PBLS was 181 mg l⁻¹, yielding low biodegradability ratios (BOD₅/COD) of 0.1. As landfill age increases, the biodegradable fraction of organic pollutants in leachate decreases due to the anaerobic decomposition occurring in the landfill site. Thus, old leachate contains much more refractory organics than young leachate. In literature, stabilized landfill leachate (age > 10 years) normally contains high quantity of NH₃-N (> 400 mg l⁻¹), moderately high strength of COD (< 4,000 mg l⁻¹), and a low BOD₅/COD ratio of less than 0.1 (Guo et al., 2010). Consequently, PBLS leachate could be classified as stabilized leachate. The concentrations of NH₃-N were also high in raw leachates at PBLS (1627 mg l-¹). The existence of high level of NH₃-N in landfill leachate over a long period of time is one of the most important problems routinely faced by landfill operators. This high quantity of unprocessed NH3-N leads to encouraged algal growth, reduced performance efficiency of biological treatment methods, accelerated eutrophication, and increased DO reduction. As a result, NH3-N is extremely toxic to aquatic organisms (Bashir et al., 2010). Obtained results showed that PBLS generates a dark black-green liquid, which could be categorized as stabilized with high concentrations of COD and NH₃-N and a low BOD₅/COD ratio (Bashir et al., 2010, Mohajeri et al., 2010a).

The values of phenols, color, suspended solids, BOD₅, COD, and NH₃-N all surpassed the allowable limits issued by Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 [Act 127]. Whereas the parameters: pH, temperature, total iron, and zinc remained within the permissible limits.

The parameters: pH, color, suspended solids, BOD₅, COD, BOD₅/COD, NH₃-N, total iron, and zinc in the current study commonly agree with the data published by previous researchers (Aziz *et al.*, 2011; Bashir *et al.*, 2010; Umar *et al.*, 2010).

3.1.3. Kuala Sepetang landfill leachate

The Kuala Sepetang landfill leachate exhibited high-intensity color and high COD (6398 Pt. Co and 1456 mg l⁻¹, respectively), which were due to the presence of organic compounds of high molecular weight (Table 1). Substantial concentration of NH₃-N (approximately 564 mg l⁻¹) was also found in raw leachate. An average BOD₅ value of 257 was recorded, and a low biodegradability ratio (BOD₅/COD) of 0.19 was obtained. The concentration of phenols, color, suspended solids, BOD₅, COD, NH₃-N, and sulfide exceeded the allowable limits issued by Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009, Malaysian Environmental Quality Act 1974 [Act 127]. While the parameters: pH, temperature, total iron, and zinc remained within the allowable limits.

The values of parameters: pH, color, suspended solids, BOD₅, COD, BOD₅/COD, NH₃-N, total iron, and zinc in Kuala Sepetang landfill leachate generally confirmed the published results (Umar *et al.*, 2010; Abu foul, 2007). Based on the reported results in literature, Kuala Sepetang landfill leachate could be considered as stabilized with relatively low refractory organic compounds, high concentrations of NH₃-N and COD (Abu foul, 2007).

3.2. Effectiveness of various scenarios in treating stabilized landfill leachate

If raw leachate is disposed without treatment, it could become a major source of water pollution because it can percolate through soils and sub-soils, causing high contamination of the receiving water. The treatment of potentially hazardous constituents of leachate prior to discharge is a legal requirement to avoid contamination of water resources to prevent both acute and chronic toxicity (Aziz et al., 2011). To reduce the negative impact of discharged leachate on environment, several techniques of water and wastewater treatment have been used. The technologies which were developed for the treatment of landfill leachate could be classified as physical, chemical, and biological (Mojiri et al., 2014; Bashir et al., 2013; Renou et al., 2008).

In general, biological treatment processes are effective for young or freshly (<5 years) produced leachate, but are ineffective for leachate from older landfills (>10 years old). In contrast, physical—chemical methods which are not favoured for young leachate treatment are advised for older leachate treatment (Mojiri *et al.*, 2014; Bashir *et al.*, 2013). Normally, the techniques are applied as an integrated system because it is not easy to achieve the satisfying treatment efficiency by using only one technology. Traditional treatment techniques generally demand multistage process treatment. To set up acceptable treatment process for removal of contaminates from leachates, various physicochemical and biological techniques and/or their different combinations could be applied. The implementation of the most suitable technique for the treatment of leachate is directly governed by the characteristics of the leachate. Leachates from different landfills vary considerably in their chemical compositions due to aforementioned factors (Renou *et al.*, 2008; Bashir et al., 2015).

Several studies were conducted in recent years to investigate the treatability of old landfill leachate generated from PBLS, as a case study in Malaysia (Table 2). From Table 2, it can be observed that old leachate treatment via biological process alone was not efficient (Aziz *et al.*, 2011). Treatment of PBLS leachate via composite adsorbent combining excellent properties of activated carbon, zeolite, rice husk carbon (RHC) and limestone was investigated. Ordinary Portland cement (OPC) was chosen to bind all adsorbents together in a single medium. Comparison study indicated that the adsorption capacity of composite adsorbent towards NH₃-N (24.34 mg g⁻¹) was higher than zeolite (17.45 mg g⁻¹) and activated carbon (6.079 mg g⁻¹) and comparable to activated carbon for COD (*Halim et al.*, 2012).

Electro-oxidation using graphite carbon electrode obtained optimal removal efficiencies for COD and color of 68% and 84%, respectively (Bashir et al., 2009). For Fenton oxidation, the experimental process achieved optimum removal values for COD and color of 58.3% and 79%, respectively (Mohajeri et al., 2010a). However, by using Electro-Fenton oxidation, the optimum removal efficiency was improved to 94.4% and 96.6% for COD and color, respectively, which revealed that Electro-Fenton process is more efficient than both Fenton and electro-oxidation (Mohajeri et al., 2010b). The performance of persulfate as sodium persulfate (Na₂S₂O₈) in treating stabilized leachate was lower than Fenton oxidation as shown in Table 2. The performance of ozone alone in stabilized leachate treatment is still low particularly in organics and ammonia removals which suggest utilizing advanced oxidant materials to improve the removal efficiency. In the case of ozone/Fenton oxidation, the optimization process achieved 78%, 98% and 20% removal of COD, color and NH₃-N, respectively at optimal operational condition of 30g m⁻³ ozone dosage, H_2O_2 0.01mol l^{-1} , Fe(II) 0.012 mol l^{-1} dosage, pH 5 and 90 min of reaction time. Consequently, the effectiveness of ozone persulfate oxidation resulted in 72%, 96% and 76% removal efficiencies of COD, color and NH₃-N, respectively, at optimal conditions of 30g m⁻³ ozone dosage, 210 min of reaction time and Persulfate dosage of 1g/1g (COD/S₂O₈²⁻ ratio). The combination of Fenton and Persulfate and advanced oxidant reagents for ozonation can improve the treatability of the stabilized leachate. Furthermore, Electro-Fenton and Ozone/Fenton oxidation processes are much more effective in removing COD and color as compared with other treatment processes (Abu Amr et al., 2013a&b). The optimum effectiveness of sequential treatment applications (cationic-anionic and anionic-cationic) was also investigated. For cationicanionic treatment, the experimentally achieved optimum removal values of color, COD and NH₃-N were 96.8%, 87.9%, and 93.8%, respectively. However, the application of the anioniccationic treatment resulted in 91.6%, 72.3%, and 92.5% removal of color, COD and NH₃-N, respectively. Consequently, the results imply that the application of the cationic-anionic sequence for the treatment of stabilized landfill leachate was more effective than the anioniccationic sequence (Bashir et al., 2011).

3

Table 2. PBLS stabilized leachate treatment efficiency using various practices

		Remo	val efficie	ncy (%)	Fir	nal concentr		
Method	Operation conditions	COD	color	NH ₃ -N	COD, mg/L	color, Pt.Co/L	NH₃-N, mg/L	References
Sequencing batch reactor (SBR)	Initial COD, 2850 mg l^{-1} ; without PAC: HRT, 2.22 day	29.2	22	71	2015	3275	405	Aziz <i>et al.</i> (2011)
Powdered activated carbon augmented SBR	Initial COD, 2850 mg l ⁻¹ ; PAC, 3g l ⁻¹ ; HRT, 2.22 day	46	31	78	1540	2895	308	Aziz <i>et al</i> . (2011)
Adsorption	5 g of composite/100 ml of leachate; composite consists of 45.94% zeolite, 15.30% limestone, 4.38% AC, 4.38% rice husk ash, and 30.00% cement used as a binder;	86		92	330	-	155	Halim <i>et al.</i> (2012)
Electro- oxidation	graphite carbon electrode, Na_2SO_4 , 1g I^{-1} ; reaction time (RT), 4h; and current density, 79.9 mA cm ⁻²	68	84	NA	600	475	NA	Bashir <i>et al.</i> (2009)
Fenton oxidation	$H_2O_2,0.033 mol/L;Fe(II),0.011moll^{-1};pH,3;andRT,145min$	58.3	79	NA	985	885	NA	Mohajeri <i>et al.</i> (2010a)
Electro-Fenton Oxidation	RT, 45 min; pH, 3.5; H_2O_2 , 0.012 mol I^{-1} ; Fe^{2+} , 0.012mol I^{-1} ; and CD, 55 mA cm ⁻² .	94.4	96.9	NA	135	130	NA	Mohajeri <i>et al.</i> (2010b)
Ozone	RT, 60min; pH, 8.5; and O_3 dosage 80 g m ⁻³	15	27	0.25	1720	2590	808	Abu Amr <i>et al</i> (2013a)

Ozone/Fenton	RT, 90 min; O_3 dosage, 30 g m ⁻³ ; Ph, 5; H_2O_2 0.01 mol I^{-1} ; and Fe^{2+} , 0.02 mol I^{-1}	78.0	99.0	20.0	445	35	648	Abu Amr <i>et al</i> (2013a)
Persulfate oxidation	RT, 240 min; pH: 8.5, COD/ $S_2O_8^{2-}$ ratio, (1g/7g); Rotation, 350rpm; and Temp. 28 $^{\circ}C$	39	55	22	1235	1600	22	Abu Amr <i>et al</i> (2013b)
Ozone/Persulfat e	RT, 210 min; O_3 dosage 30 g/m³; pH, 10; and Persulfate dosage 1g/1g (COD/ $S_2O_8^{2-}$ ratio)	72	96.0	76.0	570	140	195	Abu Amr <i>et al.</i> (2013b)
Ion exchange	Sample 100 ml; pH8.3; anion dosage 28.3cm³ followed by cation dosage of 19.6 cm³.	72.3	91.6	92.5	640	430	146	Bashir <i>et al.</i> (2011)
coagulation/floc culation	poly-aluminum chloride dosage, $1.9 \mathrm{g} \mathrm{l}^{-1}$; pH; 7.5; rapid mixing, 80rpm; time of rapid mixing, 1min; time of slow mixing, 30 rpm; time of slow mixing $_{s}$, 15min	56.76	97.26	99.18	-	835	110	Ghafari <i>et al.</i> (2010)
coagulation/floc culation)	alum dosage, 9.4 g l ⁻¹ ; pH, 7; rapid mixing, 80rpm; time of rapid mixing, 1min; slow mixing, 30rpm; time of slow mixing, 15min	84.50	92.23	94.8	-	300	305	Ghafari <i>et al.</i> (2010)

Optimum coagulant dose and pH were respectively found at 1.9 g l⁻¹ and 7.5 for poly-aluminum chloride, and 9.4 g l⁻¹ and 7 for alum, COD removal of 84.50% and 56.76% where achieved by alum and poly-aluminum chloride, respectively. Using poly-aluminum chloride, almost complete removals for physical parameters of leachate (turbidity: 99.18%, color: 97.26%, and TSS: 99.22%) were achieved; whereas alum showed inferior removal (turbidity 94.82%, color 92.23%, and TSS 95.92%). Nevertheless, results revealed poly-aluminum chloride is not as efficient as alum for COD elimination. Based on the above mentioned results, the performance of coagulation-flocculation process in stabilized leachate treatment can be considered reasonable. Nevertheless, typically, there are several drawbacks of coagulation-flocculation process including not effective for NH₃-N removal, lower removal efficiency for high-strength leachate, and production a considerable amount of sludge. Besides that, an increase in the concentration of aluminum in the liquid phase may be observed (Ghafari *et al.*, 2010).

With reference to the above mentioned subject, a preliminary treatment process such as; advances sludge process (ASP), coagulation-flocculation, Fenton oxidation (FO), and electrochemical oxidation are recommended to be used in treating stabilized landfill leachate followed by ion exchange process which can remove NH₃-N effectively.

4. Conclusions

Design, age, and other characteristics of the sanitary landfills have great effects on the quality of formed landfill leachate. Due to high concentration of contaminants in the leachate, fresh landfill leachate requires treatment prior dispose to the natural environment. Characterization of the unprocessed leachate is the fundamental step leading to the selection of efficient treatment techniques. Commonly, biological treatment methods are effective for freshly produced leachate, but are unsuccessful for the leachate treatment originated from old landfills. In contrast, physical—chemical processes which are not favored for fresh leachate treatment could be applied for old leachate treatment. Consequently, different physiochemical treatment processes conducted on leachate generated from old landfill site were highlighted in the current study. It was indicated that, a preliminary treatment process such as; advances sludge process (ASP), coagulation-flocculation, Fenton oxidation (FO), and electro- oxidation are recommended to be used in treating stabilized landfill leachate followed by another process to treat NH₃-N effectively such as ion exchange process.

References

- Abu Amr S.S., Aziz H.A, Adlan M.N. and Bashir M.J.K. (2013a), Optimization of semi-aerobic stabilized leachate treatment using ozone/Fenton's reagent in the advanced oxidation process, *J. Environ. Sci. Health, part A, Toxic/Hazar. Sub. & Env. Eng.*, **48**, 720–729.
- Abu Amr S.S., Aziz H.A, Adlan M.N.and Bashir M.J.K. (2013b). Pretreatment of stabilized leachate using Ozone/Persulfate oxidation process, *Chemical Engineering Journal*, **221**, 492–499
- Abu Foul A.E.M. (2007), Physical-chemical treatment of stabilized leachate using limestone, activated carbone and zeolite. PhD thesis. School of Civil Engineering, University Sains of Malaysia (USM), Malaysia.
- Ahn S., Congeevaram S., Choung Y.K. and Park J. (2008), Enhanced phenol removal by floating fungal populations in a high concentration phenol–fed membrane bioreactor, *Desalination*, **221**, 494–501
- APHA. (2005), Standard Methods for the Examination of Water and Waste Water. 21st edition. American Public Health Association, Washington, DC.
- Aziz S.Q., Aziz H.A., Yusoff M.S. and Bashir M.J.K. (2011), Landfill leachate treatment using powdered activated carbon augmented sequencing batch reactor (SBR) process: Optimization by response surface methodology, *Journal of Hazardous Materials*, **189**, 404-413.

Aziz S.Q., Aziz H.A., Yusoff M.S., Bashir M.J.K. and Umar M. (2010), Leachate characterization in semi-aerobic and anaerobic sanitary landfills: A comparative study, *Journal of Environmental Management*, **91**, 2608-2614.

- Bashir M.J.K., Isa M.H., Kutty S.R.M., Awang Z.B., Aziz H.A., Mohajeri S. and Farooqi I.H. (2009), Landfill leachate treatment by electrochemical oxidation, *Waste Management*, **29**, 2534-2541.
- Bashir M.J.K., Aziz H.A., Yusoff M.S. and Adlan M.N. (2010), Application of response surface methodology (RSM) for optimization of ammoniacal nitrogen removal from semi aerobiclandfill leachate using ion exchange resin. Desalination, **254**, 154 161.
- Bashir M.J.K., Aziz H.A.,and Yusoff M.S. (2011). New sequential treatment for mature landfill leachate by applying cationic/anionic and anionic/ cationic processes: optimization and comparative study. *Journal of Hazardous Materials*, **186**, 92-102.
- Bashir M.J.K., Aziz H.A., Aziz S.Q.and Abu Amr, S.S. (2013). An Overview of Electro-oxidation Processes Performance in Stabilized Landfill Leachate Treatment. *Desalination and Water Treatment*, **51**, 2170–2184.
- Bashir M.J.K., Aziz H.A., Abu Amr S.S., Sethupathi S., Ng C.A. and Lim J.W. (2015). The competency of various applied strategies in treating tropical municipal landfill leachate. *Desalination and Water Treatment*, **54**, 2382-2395.
- Davis M.L. and Cornwell D.A., (2008), Introduction to Environmental Engineering. 4th edition. (International Edition). McGraw Hill, New York.
- El-Fadel M., Bou-Zeid E., Chahine W. and Alayli B. (2002), Temporal variation of leachate quality from presorted and baled msw with high organic and moisture content, *Waste Management*, **22**, 269–282.
- Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill)
 Regulations.under the Laws of Malaysia-Malaysia Environmental Quality Act 1974 (2009)
- Ghafari S., Aziz H.A.and Bashir M.J.K. . (2010). The use of poly-aluminum chloride and alum for the treatment of partially stabilized leachate: A comparative study, *Desalination*, **257**,110-116.
- Guo J.S., Abbas A.A., Chen Y.P., Liu Z.P. and Chen P. (2010), Treatment of landfill leachate using a combined stripping, Fenton, SBR, and coagulation process, Journal of Hazardous Materials, **178**, 699 –705.
- Halim A.A., Aziz H.A., Johari M.A.M. Ariffin K.S. and Bashir M.J.K. (2012). Semi aerobic Landfill Leachate Treatment Using Carbon-Minerals Composite Adsorbent, *Environmental Engineering Science*, **29**, 306-312.
- Kiayee S.B (2013), Impact of Municipal Waste Leachate Application on Soil Properties and Accumulation of Heavy Metals in Wheat (*Triticum aestivum* L), *International Journal of Scientific Research in Environmental Sciences*, **1**, 1-6.
- Kujawski W., Warszawsk A., Ratajczak W., Porebski T., Capata W. and Ostrowska I. (2004), Removal of phenol from wastewater by different separation techniques, *Desalination*, **163**, 287–296.
- Kurata Y., Ono Y. and Ono Y. (2008), Occurrence of phenols in leachates from municipal solid waste landfill sites in Japan, *Journal of Material Cycles and Waste Management*, **10 (2)**, 144-152.
- Matsufuji Y., Hanashima M., Nagano S. and Tanaka A. (1993), Generation of Greenhouse Effect Gases from Different Landfill Type, *Engineering Geology*, **34**, 181-197.
- Mohajeri S., Aziz H.A., Isa M.H., Bashir M.J.K., Mohajeri L., and Adlan, M.N. (2010a). Influence of Fenton reagent oxidation on mineralization and decolorization of municipal landfill leachate. *Journal of Environmental Science and Health, Part A/Toxic/Hazardous Substance & Environmental Engineering*, 45, 692-698.
- Mohajeria S., Aziz H.A, Isa M.H., Zahed M.A. and Adlan M.N. (2010b), Statistical optimization of process parameters for landfill leachate treatment using electro-Fenton technique, *Journal of Hazardous Materials*, **176**, 749–758.
- Mojiri A., Aziz H.A. and Aziz S.Q. (2013), Trends in Physical-Chemical Methods for Landfill Leachate Treatment, *International Journal of Scientific Research in Environmental Sciences*, **1**, 16-22.

- Mojiri A., Aziz H.A., Zaman N.Q., Aziz S.Q. and Zahed M.A. (2014), Powdered ZELIAC augmented sequencing batch reactors (SBR) process for co-treatment of landfill leachate and domestic wastewater, *Journal of Environmental Management*, **139**, 1-14.
- Qasim S.R. and Chiang W. (1994), Sanitary landfill leachate: generation. Technomic Publishing, Lancaster, Control and treatment.
- Rafizul I.M. and Alamgir M. (2012), Characterization and tropical seasonal variation of leachate: Results from landfill lysimeter studied, *Waste Management*, **32**, 2080–2095.
- Renou S., Givaudan G.J., Poulain S., Dirassouyan F. and Moulin P. (2008), Landfill leachate treatment: Review and Opportunity, *Journal of Hazardous Materials*, **150**, 468-493.
- Tziotzios G., Economou C.N., Lyberatos G. and Vayenas D.V. (2007), Effect of the specific surface area and operating mode on biological phenol removal using packed bed reactors, *Desalination*, **211**, 128–137.
- Umar M., Aziz H.A. and Yusoff M.S. (2010), Variability of parameters involved in leachate pollution index and determination of LPI from four landfills in Malaysia, *International Journal of Chemical Engineering*, Volume 2010, Article ID 747953, DOI:10.1155/2010/747953, 1–6.
- Uygur A. and Kargi F. (2004), Biological nutrient removal from pre-treated landfill leachate in a sequencing batch reactor, *Journal of Environmental Management*, **71**, 9-14.
- Xiao M., Zhou J., Tan Y., Zhang A., Xia Y. and Ji L. (2006), Treatment of highly–concentrated phenol wastewater with an extractive membrane reactor using silicone rubber, *Desalination*, **195(1-3)**, 281–293.
- Yamamoto O. (2002), Solid waste treatment and disposal experiences in Japan. Proceedings of International Symposium on Environmental Pollution Control and Waste Management (EPCOWM'2002), Tunis, 417-424.