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Comparison and Evaluation of Different Leachate Treatment Processes for Chemical Oxygen Demand and Color Removals - Statistical Assessment

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Abstract

The performance of seven oxidation treatments on stabilized landfill leachate namely; O_3, Fenton, Fenton followed by O_3, persulfate, persulfate followed by O_3, simultaneous O_3/Fenton, and simultaneous O_3/persulfate were evaluated and compared. The study aims to assess the variation between all the seven oxidation treatments with regards to COD and color removal. The analysis of variance (ANOVA) and Tukey's test showed that there were significant differences between the performances of the treatments. The O_3 treatment removed the lowest COD and color while the simultaneous O_3/Fenton, Fenton followed by O_3, simultaneous O_3/persulfate and persulfate followed by O_3 removed the highest. Hence, these treatments are recommended to remove organics from stabilized landfill leachate.

Keywords: Stabilized leachate; Treatment; Oxidation; Analysis of variance (ANOVA); Tukey's test.

1. Introduction

Municipal solid waste comprises of domestic and commercial waste, construction debris, and sanitation residue (Agamuthu and Fauziah, 2011). Landfilling is one of the most economical methods to dispose these waste (Tan *et al*., 2013; Bashir *et al*., 2018). Ninety percent of the capacity of the landfills are used to discard the solid waste. This large volume of solid waste can contaminate the underlying soil and seep into the groundwater (Shabiimam and Dikshit, 2012).

The landfill leachate contains several contaminants that could potentially cause detrimental health effects, either due to direct contact (e.g., skin damage) or through drinking polluted water or eating contaminated crops (EPA, 2007). Moreover, soil and agricultural productivity can decrease if leachate-polluted water is used for irrigation (Alslaibi *et al*., 2010).

Stabilized leachate, as indicated by a low BOD_5/COD ratio (i.e., low biodegradability) is unsuitable for to be treated biologically. Therefore, additional physicochemical processes are necessary for the pre-treatment and post-treatment of leachate (Tauchert *et al*., 2006). Thus, before discarding leachate into the environment, dedicated treatment facilities are required (Goi *et al*., 2009). Considerable attention has been given to advanced oxidation processes to reduce the organic load in leachate. Recently, several applications and techniques have been recommended to treat stabilized leachate such as persulfate, persulfate combined with H2O2 and $AlSO_4$, ozonation (O_3), Fenton, photo-Fenton, electro-photo-Fenton, ion exchange and adsorption isotherm (Abu Amr *et al.*, 2017; Hilles and Abu Amr 2016; Mohajeri *et al.*, 2010; Abu Amr *et al.*, 2013 a & b; Bashir *et al.*, 2010 & 2011). Although the performances of these applications recorded higher removal for organics from stabilized leachate, high variations in the removal efficiencies have been reported. Hence, this study aimed to evaluate and compare the efficiencies of different oxidation processes on stabilized landfill leachate. In this study, the performance of seven oxidation processes for stabilized leachate namely; ozonation, Fenton reaction, ozone/Fenton, Fenton followed by ozone, persulfate oxidation, ozone/persulfate, persulfate followed by ozone was evaluated and compared. Their performances to remove COD and color from leachate were statistically analyzed using the Analysis of Variance (ANOVA) and Tukey's test to identify the sources of differences between the different oxidation processes.

2. Material and methods

2.1 Site Location and Characteristics

The Pulau Burung Landfill Site (PBLS), is located in Byram Forest Reserve, in the town of Nibong Tebal, Pulau Pinang, Malaysia at a latitude of 5 12'03"N and longitude of 100°25'24"E with a total area of 63 ha (Mohajeri, 2010). The satellite image of the PBLS is shown in Figure 1. PBLS is equipped with a natural marine clay liner and three leachate collection ponds (Bashir *et al.*, 2011). PBLS satellite image is shown in Figure 1. The leachate collected from PBLS is considered as a semi-aerobic stabilized leachate. In the first ten years of operation, PBLS did not manage to control the leachate. However, in 1990, PBLS became a semi-aerobic system Level II MSW Sanitary Landfill and was upgraded to a Level III Sanitary Landfill in 2001. PBLS is the only operating MSW landfill in Pulau Pinang. The major waste components are food waste, paper, and plastics (Aziz *et al.*, 2010; Omran *et al.*, 2009). These municipal wastes are dumped and compacted by bulldozers before covered with soil. Leachate generated is collected through a leachate collection pipe and flows into detention ponds. These ponds are aerated intermittently via diffused aeration with a high-pressure pump.

Figure 1. Satellite image of PBLS. Imaging date (12/10/2018)

Table 1. Characteristics of stabilized landfill leachate collected from PBLS

Parameters	Jan. 2011 – June. 2012 (9 Samples)		Standard*
	Value	Average	
COD (mg/L)	1780 - 2530	2025	400
BOD_5 (mg/L)	70 - 107	93	----
BOD_5/COD	0.034 – 0.05	0.043	----
NH_3-N (mg/L)	790 - 1170	915	5
Color (PL Co.)	3450 - 4530	4198	100
pH	8.4 – 8.9		6 – 9
Suspended solids (mg/L)	186 - 225	197	50
Conductivity, (μS/cm)	18325 - 20350	19,430	-----
Fe^{2+} (mg/L)	6.7 - 12	9	5
SO_4 (mg/L)	165 - 202	187	500 – 1000**

*Malaysia Environmental Quality regulation 2009, Control of pollution from the solid waste transfer station and landfill, Act 1974.
**National standards for Environmental quality 2001, Waste water treatment guidance manual, Ministry of Syrian Environment

2.2 Sampling

Leachate samples were collected from a leachate aeration pond of a semi-aerobic stabilized landfill leachate at the PBLS. Eight samples were collected during the period between June 2012 and December 2013. Approximately 20 L of leachate was manually collected each sampling time and placed in plastic containers. The samples were immediately transported to the laboratory, characterized, and cooled at 4 °C to minimize the biological and chemical reactions. The Standard Methods for the Examination of Water and Wastewater (APHA, 2005) were used to collect and preserve the samples. General characteristics of leachate sample was presented in table 1.

2.3 Experimental procedure

2.3.1 Ozone Oxidation

The O_3 oxidation experiments on the leachate sample were performed in a reactor. The height and internal diameter of the reactor were 65 cm and 16.5 cm, respectively. An ozone generator (model BMT 803, BMT Messtechnik, Germany) produced the O_3 needed for the experiment. The O_3 concentration was measured using an ultraviolet gas ozone analyzer (model BMT 964, BMT, Messtechnik, Germany). The analyzer has a measurement range of 30-80 g/m^3. To determine the optimal initial pH in treating the stabilized leachate, the initial pH of the leachate samples was adjusted between 3 and 11. The optimal reaction time was also determined by varying the reaction time between 10 and 120 min (Abu Amr *et al.*, 2013a; Tizaoui *et al.*, 2007).

2.3.2 Ozone/Fenton oxidation

The effect of the Fenton molar ratio, Fenton concentration, pH, and reaction time was investigated to determine optimal conditions for the leachate treatment. Initial leachate pH was adjusted to the desired value with the use of 7 to 10 mL of 5 M sulfuric acid or 5 M sodium hydroxide. During the experiment, the leachate was continuously stirred with a magnetic stirrer. All experiments were performed at room temperature and atmospheric pressure. Fenton reagent (H_2O_2/Fe^{2+}) was combined with the ozonation process to treat the leachate sample. After each experiment, the samples were immediately withdrawn to avoid iron precipitation in the reactor. The samples were then allowed to settle for 30 min at higher pH (>8) before analysis to quench the reaction and improve settling of iron sludge (Mohajeri *et al*., 2010). To evaluate the synergistic effects of combined ozone and Fenton reagent as one reaction process, the treatment efficiency of the $O_3/H_2O_2/Fe^{2+}$ system was compared with other combination of treatments (i.e., O_3 alone, Fenton oxidation alone, Fenton oxidation followed by O_3). Finally, the results of the above three oxidation processes was compared with the simultaneous O_3/Fenton (Abu Amr *et al*., 2013b).

2.3.3 Ozone/persulfate oxidation

The effectiveness of the combined O_3 and persulfate ($O_3/S_2O_8^{2-}$) treatments were also investigated. Persulfate ($Na_2S_2O_8$, M = 238 g/mol) was mixed with the leachate sample during the ozonation process as one reaction step. The $COD/S_2O_8^{2-}$ ratio (g/g), pH, and reaction time during the ozonation process were evaluated under optimal operation conditions. To evaluate the synergistic effects of combined O_3 and persulfate as one reaction process, the treatment efficiency of the $O_3/S_2O_8^{2-}$ system was compared with other treatment applications (such as O_3 alone, persulfate oxidation alone, as well as persulfate oxidation followed by O_3) (Abu Amr *et al*., 2013).

2.4 Statistical analysis

The effect of different leachate treatment on COD and color removals was studied and analyzed by one-way analysis of variance (ANOVA) to decide whether different treatment give the same percentage of COD and color removal or not (Talebi *et al*., 2014 and 2015). The significance level of 0.05 is used to make a decision (The P value is the probability of finding a more extreme, results when the null hypothesis (H_0) is true. If p- value is less than 0.05, the null hypothesis will be rejected, and fail to reject the hypothesis if more 0.05). The results were further analyzed by Tukey's test to identify the treatments that give different COD and color removals. R statistical software was used for analyzing the data.

2.5 Analytical study

The Standard Methods conducted all tests for the Examination of Water and Wastewater (APHA, 2005). The concentrations of COD, color, NH_3-N, SS, Fe^{2+}, and SO_4 were determined using HACH model 2800 Spectrophotometer. pH was measured on site using a portable digital pH/mV meter (WITEG, W-100, Germany). The Electrical Conductivity (EC) was measured as μS/cm using a portable multi-purpose (Multi 340i, Germany). HACH instruments were pre-celebrated before use, and standard solutions for COD and NH_3-N were used to determine the accuracy of the measurements. As the maximum range of the parameter in HACH is limited and the parameters concentration high; Samples were diluted from 1 to 5 times for COD, color, and other parameters, while for NH_3-N, the sample was diluted to 1000 time because of vary low range (2.5 mg/L) and high initial concentration (> 1000 mg/L). The desired program number and wavelength were selected; a high range program was used for COD color and NH_3–N as a suitable range of wastewater with high concentrations.

3. Results and discussion

The characteristics of the landfill leachate sample used for the ozone oxidation treatment are shown in Table 1. It can be seen that COD and NH_3-N values of the landfill leachate were quite high. This leachate was categorized as old with very low biodegradability (BOD_5/COD = 0.034 to 0.050) and very high color intensity (5,320 Pt-Co) which cause a high level of NH_3–N (1,170 mg/L) in this landfill leachate, the high level of COD and NH_3-N has been one of the most important problems faced by landfill operators over a long period. The high quantity of unprocessed NH_3–N depletes dissolved oxygen in a process known as eutrophication (Bashir *et al*., 2010). Aziz *et al.* (2007) indicated that the high color value in landfill leachate is caused by the large quantity of organic substances. The concentrations of heavy metals in PBLS were low: zinc (0.1 mg/L to 1.8 mg/L), copper (0.1 mg/L to 0.4 mg/L), manganese (0.6 mg/L to 1.1 mg/L), cadmium (<0.4 mg/L), and iron (0.32 mg/L to 7.5 mg/L) (Aziz *et al*., 2007). Different leachate treatment processes were thought to affect the percentage of COD and color removals. An experiment was carried out to compare the performance of seven different leachate treatment processes namely: Ozone, persulfate, Fenton, combined Ozone/ Fenton, combined Ozone/persulfate, ozone followed by Fenton and ozone followed by persulfate regarding COD and color removals. The average percentage of COD and color removals are presented in a bar chart (Fig.2a and b) showing the percentage of removal for each leachate treatment. It can be seen that the differences in the COD and color removals for all selected leachate treatment. The results of the percentage of COD and color removals obtained from different experiments were analyzed by analysis of variance (ANOVA). The results of ANOVA (Table 2) showed a significant difference in the percentage of COD and color removals for the selected leachate treatment processes. The data were further analyzed by Tukey's test for multiple comparisons to identify the differences in COD and color removals among different leachate treatments, The results of Tukey's test for COD removal showed a significant difference (p-value < 0.001) between:

O_3 and Fenton
persulfate and Fenton
O_3 and simulatenous O_3/Fenton
persulfate and simulatenous O_3/Fenton
Fenton followed by O_3 and O_3 alone
persulfate followed by O_3 and O_3 alone
persulfate and O_3
simulatenous O_3/persulfate and O_3
persulfate and simulatenous O_3/Fenton
persulfate and persulfate followed by O_3
simulatenous O_3/persulfate and persulfate

other combinations did not show a significant difference at p-value < 0.001. While, the results of Tukey's test for color removal exhibited different behavior since only three treatments did not show a significant difference regarding color removal, the three treatments are O_3/ Fenton-Fenton+ Ozone, O_3/Persulfate-Fenton+ Ozone and Persulfate + O_3-O_3/Persulfate. A significant difference between ozone alone and other oxidation processes could be attributed to the high concentration of organics in leachate containing a considerable amount of dissolved ozone in the aqueous phase from the beginning of the reaction (Rivas *et al.*, 2003).

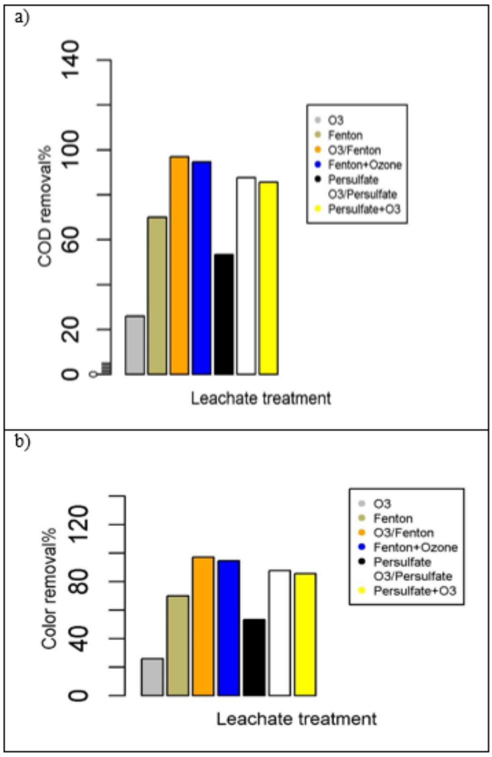

Figure 2. Showing the behaviour of selected treatment on a) COD removal and b) Color removal.

The performance of ozone alone for COD and color removal is very weak as shown Figure1 (COD= 15.67% and color = 26%). The Fenton oxidation is more efficient for leachate treatment than O_3 alone. The performance of ozone after Fenton treatment did not show improvement in terms of COD and color removals; this result is in agreement with the results reported by Goi *et al.* (2009). The highest removal efficieny was obtained by using simultaneous ozone/Fenton oxidation ($O_3/H_2O_2/Fe^{2+}$). In the O_3/Fenton oxidation, the cooperative reaction of ozone and Fe^{2+} ions with H_2O_2 was performed at natural pH (7) (Abu Amr and Aziz 2012) to form hydroxyl radical (Eqs. 1&2). Moreover, H_2O_2 was dissolved in water and dissociated into hydro peroxide ion (HO^{2-}), which rapidly reacted with the ozone to initiate a radical chain mechanism that generates hydroxyl radicals.

Table 2. The results of ANOVA for the effect of treatment time on COD and color removals

	Df	Sum Sq	Mean Sq	F value	Pr(>F)
			COD removal		
Treatment	6	5201	866.8	54.83	< 32e-09
Residuals	14	221	15.8		
Signif. codes	0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1				
			Color removal		
Treatment	6	12073	2012.1	556.7	< 96e16
Residuals	14	51	3.6		
Signif. codes	0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1				

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH\cdot \quad (1)$$
$$H_2O_2 + 2O_3^2 \rightarrow OH\cdot + 3O_2 \quad (2)$$
$$HO_2^- + O_3 \rightarrow OH_2\cdot + O_3 \quad (3)$$

Tizaoui *et al.* (2007) used H_2O_2 for improving ozone efficiency in landfill leachate treatment, as well as for improving the removal efficiencies for COD (from 27% to 50%) and color (from 87% to 94%). When the Fenton process was used, the obtained removal rates for COD and color were 58.1% and 78.3%, respectively (Mohajeri *et al.*, 2010).

Persulfate oxidation is more efficient for leachate treatment than O_3 alone. Although the performance of O_3 after $S_2O_8^{2-}$ is improved, the removal efficiency is also improved by the advanced oxidation system (O_3/ $S_2O_8^{2-}$). Persulfate oxidation can be enhanced by the release of sulfate radicals, which have powerful effects on the oxidation of organics (Kolthoff *et al.*, 1947; Watts *et al.*, 2001). The generation of sulfate radicals during oxidation can be significantly enhanced by catalysts, such as heat and UV radiation (Eq. 4 - 6), which were found to improve the persulfate oxidation potential (Gao *et al.*, 2012; Langlais *et al.*, 1991).

$$S_2O_8^{2-} + 2H^+ + 2e^- \rightarrow 2HSO_4\cdot^- \quad (4)$$
$$SO_4^{2-} + OH\cdot \rightarrow SO_4\cdot^- + OH^- \quad (5)$$
$$S_2O_8^{2-} + \text{Thermal activation} \rightarrow SO_4\cdot^- + SO_4\cdot^- \quad (6)$$
$$(30^oC < T < 90^oC)$$

Shiying *et al.* (2009) initiated sulfate radical by activating persulfate, using Microwave (MW)–activated persulfate oxidation. The degradation of organics evaluated the effectiveness of the process in wastewater; 83% to 95% of COD removal was obtained. Ozone can be used in initiating sulfate radical formation from persulfate during the ozonation process and improve the oxidation potential. Although the following processes namely Ssimultaneous O_3/Fenton, Fentone+Ozone, simultaneous O_3/persulfate and persulfate+O_3 showed the highest removal for COD and color, however O_3/persulfate can be considered as the most suitable process for leachate treatment because of O_3/persulfate does not produce sludge after the treatment process. One of the main environmental problems in O_3/Fenton process is the iron sludge production the final effluent discharge, which requires additional appropriate management prior to discharge.

4. Conclusion

The current study evaluated and compared the performance of different oxidation processes for stabilized leachate treatment. It can be said that ozone alone has the lowest treatment efficiency compared to other treatment processes. Furthermore, persulfate alone and Fenton alone showed a significant improvement in removing COD and color removals compared with ozone alone. Simultaneous O_3/Fenton, Fentone+Ozone, simultaneous O_3/persulfate and persulfate+O_3 showed the highest removal which can be used for treatment processes to remove organics from stabilized landfill leachate. Furthermore, O_3/persulfate can be considered as the most suitable process for leachate treatment because of O_3/persulfate does not produce sludge after the treatment process compared with O_3/Fenton and Fenton followed by O_3.

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