Leachate treatment using a novel sustainable fixed bed based method

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Abstract

Toxic and environmentally damaging leachate is a product of municipal solid waste disposal in landfill systems. Currently, 51% of landfill leachate (LFL) produced in Irish landfill sites is discharged directly into sewer mains with 48% being treated in increasingly overloaded regional wastewater treatment plants. These discharge and treatment options are inadequate, costly and pose risks for both public and environmental health. Unlike other EU countries onsite treatment of leachate in Ireland is uncommon (<1%), but could represent a viable and sustainable alternative to current practices. The current study has shown that low cost adsorption material, such as oyster shells and pumice, are capable of reducing the concentration of ammonia, phosphate and nitrate from these waste streams. In addition, microbial isolates from leachate have demonstrated the ability to reduce toxic compounds, such as ammonia and phosphate. This research aims to combine both, adsorption and bioremediation into single treatment process using fixed bed systems. This treatment should reduce LFL to within acceptable limits set by the EPA (Ireland) for the discharge of leachate into receiving bodies. This treatment option will also be low-cost and have the ability to be implemented onsite in Irish landfills.

Keywords: Landfill leachate, Novel Treatment, Wastewater, Bioremediation, Adsorption

1. Introduction

The generation of heavily polluted leachate constitutes a major drawback of Municipal Solid Waste (MSW) landfills. While there has been a decline in the number of landfills over recent years, the generation of leachate is a legacy problem and its treatment is a major management issue for landfills operators within the European Union (EU) (Zhang *et al*., 2009; Brennan *et al*., 2016, 2017). LFL is defined by McCarthy *et al.*, (2010) as 'liquid, which has percolated through the waste, picking up suspended and soluble materials that originate from or are products of the degradation of the waste'. As liquid penetrates through the solid matrix it assists with biochemical, chemical and physical reactions, directly influencing the quality and quantity of the leachate produced (Kamaruddin, 2015). Many methods are used to treat LFL, however, most are adapted from wastewater treatment methods (Raghab *et al.*, 2013). Usually a combination of both biological and physiochemical methods are effective, as it can be difficult obtain satisfactory results with just one method due to the variety of composition between different LFL (Kargi and Pamukoglu, 2004). For example, LFL that has a high organic content is best treated biologically, whereas LFL with a low organic content is best treated physicochemically (Kheradmand *et al.*, 2010). A number of treatment options have been successfully employed to treat LFL. A study carried out by Paskuliakova *et al.*, (2016) applied chlorophytes to reduce the total ammonia nitrogen and total organic nitrogen. Zayen *et al*., (2016) combined processes of anaerobic digestion, lime precipitation microfiltration and reverse osmosis to treat LFL, while Kaur *et al*., (2016) used cow-dung ash as an adsorbents material to assess for the removal of organic material. Even though these treatments have been successful, it is important to investigate other option, especially those that are low cost and can be implemented onsite in Irish landfills. Currently, in Ireland, over fifty urban wastewater treatment plants (WWTP's) receive and treat MSW LFL, requiring transport and costly aerobic biological treatment. Volumes and composition of LFL collected at these sites varies greatly depending on the content, size and age of the specific landfill (McCoole *et al*., 2010). In 2013 there was approximately 1.1 million m^3 of LFL collected in Ireland, which was either discharged directly to sewers (51%) or tankered to WWTP's (48%) for final treatment, with only 1% receiving any onsite treatment (EPA, 2015). Currently, over fifty WWTP's receive leachate from MSW landfills. Treatment of leachate in WWTP's is not effective, as the systems employed in these treatment centres are often inadequate and do not effectively treat leachate to the discharge limits. Another drawback for WWTPs is the stringent emission limits. Non-compliance with ammonia and total nitrogen emission values in WWTP's has been attributed to leachate loading at these plants, resulting in the discontinuation of leachate acceptance by WWTP's. This has resulted a 30% decrease in the number of WWTP's treating leachate from 2010 to 2015 (McCarthy *et al*., 2010; EPA, 2015; Brennan *et al*., 2016). For this reason, it is of economic and environmental importance to investigate the best way to treat LFL, in order to develop a cost effective, suitable treatment, that will ultimately reduce LFL consitiuients to required discharged limits. The main purpose of this study is to combine both biological, in terms of bioremediation, and the physicochemical treatment of adsorption, into a novel cost effective system to treat LFL. This study utilised low cost adsorption material, and bacteria isolated from leachate to treat LFL from an Irish landfill. Both treatment processes where combined into a continuous column system. The main aims of the study were to; (1) evaluate the effectives of the column system in terms of overall % removal and, (2) to treat LFL to discharge limits set by EPA, Ireland for the discharge of wastewater to receiving bodies.

2. Materials and methods

2.1 Site description and leachate collection

LFL used in this study was sourced from Powerstown Landfill, Co. Carlow, Ireland. The landfill is located 8 km south east of Carlow Town in a rural setting and has been operational since 1975. The site consists of three different phases; phase 1 (P1) which operated from 1975-1990, phase 2 (P2) which operated from 1991-2006, and phase 3 (P3) opened in 2006 and is due to close before January 2018. P3 consists of four lined cells, surface water settlement pond, leachate tank and green waste composting area. Leachate collection systems are in operation in both P3 and P2. It was decided to use LFL generated in P3 as it is currently in operation and generates a more concentrated leachate than the other phases. LFL samples were collected in January and February 2017 and stored at 4°C until use within 48 hrs.

2.2 Continuous column system and operation

Three sequential PVC columns (11 cm diameter, 30 cm height, and IC 2850 cm^3) were utilised in this study. The first column (C1) was packed with c. 1 kg of soil to a height of 20 cm. The column was then spiked with a 500 ml overnight broth culture of 20 previously isolated leachate degrading microorganisms in nutrient broth. The soil/microbe mixture was left to incubate for 48 hrs at room temperature (25°C) after which the liquid was allowed to drain off. The second column (C2) was packed with c. 1.3 kg of crushed oyster shells to a height of 20 cm. The final column (C3) was packed with 650 g of pumice stone to a height of 20 cm. Both adsorption materials were prepared by washing with deionised water and drying at 100°C for 24 hr. Before commencement of the experiment deionised water was washed through the column in a down flow direction to withdraw trapped air between the materials. LFL was actively pumped into C1 at a flow rate of 5 mls/min and allowed to filter via gravity into C1 and C2 sequentially. The total active volume and retention time of combined system was 2.1kg and 3 hrs, respectively.

2.3 Leachate analysis

Leachate was analysed before and after treatment in each column. Ammonia and BOD were analysed according to standard method (APHA 2008). COD, phosphates and nitrates where analysed with HACH DR6000, using HACH COD vials, Nitraver5 pillows and phosphate reagent.

3. Results and discussion

3.1 C1 –Bioremediation

C1 consisted of soil inoculated with a master mix of 20 microbes that were isolated from LFL sourced from Powerstown Landfill in November 2015. All of the selected strains have displayed the ability to utilise leachate, as well as, heavy metals, ammonia, phosphate and nitrates as a sole carbon source (unpublished data). Leachate applied to C1 varied in composition, depending on its collection date. Laboratory analysis of leachate composition corresponded to the data received from Powerstown, indicating that P3 is c. 10 years old (Table 1). Table 1 also shows the general composition of leachate from Powerstown over the past 8 years and the EPA discharge limits to release treated leachate into water bodies. These results are comparable to leachate previously studied from an intermediate landfill (5-10 years) (Renou *et al.*, 2008). The results from column 1 indicated that ammonia, phosphate and nitrate removal efficiencies were 89%, 41% and 44%, respectively (Fig.1). Despite this substantial reduction, specifically for ammonia, none of the parameters measured reached discharge limits after treatment in C1, with final. effluent concentrations of 108 mg/l of ammonia, 2.75 mg/l of phosphate and 82 mg/l of nitrates. However, it should be noted that the HRT of C1 was 90 min and % removal efficiencies may be improved by increasing this to allow more contact time between the microbial consortia and the influent. In addition, the application of effluent recycling within C1 could further improve bioremediation of these constituents.

3.2 C2- Adsorption 1

Fixed bed adsorption has become a frequently used application in wastewater treatment processes. The performance of packed beds is described through the concept of the breakthrough curve. The breakthrough curve is used to evaluate the performance of the column system. From this curve it is possible to work out the overall removal capacity of the columns (Rao and Viraraghavan, 2002; Aksu and Gönen, 2004) .

Table 1 Leachate composition form Powerstown landfill over the past 8 years, compared with leachate used in this study and EPA discharge limits in Ireland

Compounds	Ranges from Powerstown Landfill years 2009-2015	Leachate used in this study	EPA Limits
Ammonia (mg/l N)	360-960	790- 1010	≤4
BOD (mg/l O_2)	46-180	112-170	≤5
COD (mg/l O_2)	539-2710	450- 650	≤40
Nitrate (mg/N)	Not measured	89-120	≤50
Phosphate (mg/P)	1.2-6.1	3.6	≤0.4

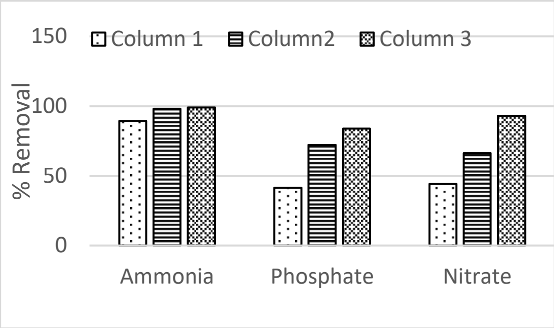

Figure 1 Overall % removal of each column for Ammonia, Phosphate and Nitrates

The total adsorbed ions, can be obtained by the integrating the plot of the adsorbed concertation (C_{ad}) versus the flow time (t). This plot is needed to determine the $q_{total.}$ This is the total amount of ions that have been removed from the leachate.

$$q_{total\ (mg)} = \frac{QA}{100} = \frac{Q}{1000}\int_{t=0}^{t=t_{total}} C_{ad}dt$$

The total amount of ions delivered to the system (m_{total}) is determine by the following:

$$m_{total}\,(mg) = \frac{C_o Q t_{total}}{1000}$$

In this equation the Q and the t_{total} represent the flow rate (ml/min) and the total flow time (min). Both of these equation are required to evaluate the removal efficiency of the column. The equations make up the total removal as a percentage:

$$Total\ removal\ (\%) = \frac{q_{total}}{m_{total}} x\ 100$$

Oyster shells were chosen for the adsorption material of C2, as readily available waste product of the aqua industry within Ireland. Results for C2 showed it was particularly effective in reducing ammonia with a ≥98% removal efficiency (Table 2). As such, ammonia was reduce to 25 mg/l in C2 effluent of C1. Increased ammonia removal is highly desirable as elevated concentrations are the main reason why WWTP's often refuse to accept LFL. Elevated ammonia concentrations can put stress on the stringent discharge limits for treated wastewater (≤4 mg/l). C2 has also shown the ability to reduce phosphate and nitrates. Although, neither were reduced below discharge limits, nitrates were significantly close at 52.23 mg. l (discharge limit; ≤50 mg/l). The final effluent concentration of C2 alongside the overall removal efficiencies are recorded in Table 2 .

3.3 C3- Adsorption 2

C3 contained pumice stone, which is a low cost adsorption material. The aim of this column was to act as a fine filter for the removal of the final amounts of ammonia, phosphate and nitrate. Over a 9 hr period, all three compounds were treated to within discharge limits (Table 3). Figure 2 shows the final effluent concentration for phosphate and nitrate.

3.4 Overall system

In general, results of the combined system were good with the discharge limits set by the EPA for the compounds investigated being met in the final effluent over a short time frame (c. 10 hrs) (Table 3). It can seen (Table 3) that

Table 2 Column 2 final effluent concertation and overall % removal

	C_o (mg.L)	Q (mls.min)	Total % Removal	Q_{eq} (mg.g)	Effluent Conc. (mg.l)
Ammonia	1040	5	98.10	0.602	24.65
Phosphate	5.26	5	72.1	0.0054	1.18
Nitrate	187.3	5	66.50	0.172	52.23

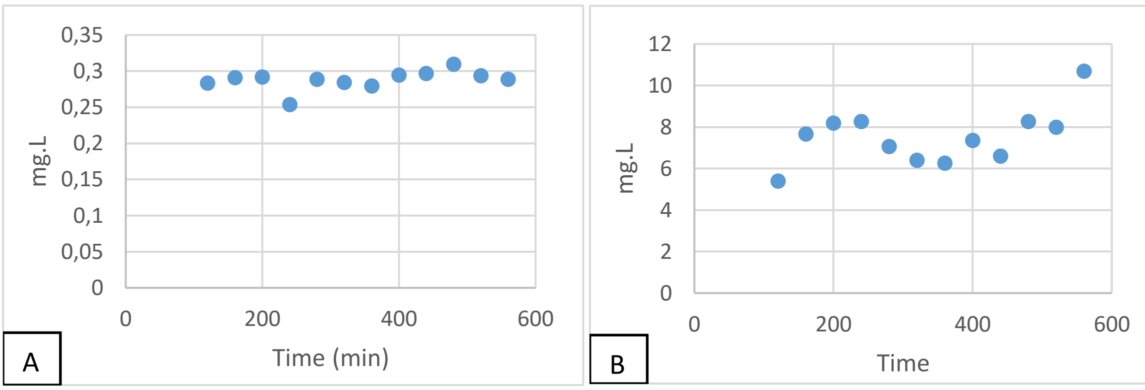

Figure 2 (A) Phosphate effluent concertation and (B) Nitrate effluent concertation

Table 3 Column 3 final effluent concertation and overall % removal

	Co (mg.l)	Q(mls.min)	Total % Removal	Q_{eq}(mg.g)	Effluent Conc. (mg.l)
Ammonia	428	5	98.95	0.62	2.3
Phosphate	5.26	5	84.78	0.0067	0.28
Nitrate	187.33	5	92.01	0.266	4.2

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Table 4 Results of combined system

	Co (mg/l)	Q (mls/min)	Total % Removal	Qeq (mg/g)	ORL (m^{-3} d^{-1})	Effluent Conc. (mg/l)	EPA Limits (mg/l)
Ammonia	428	5	98.93	0.648	2.04	2.3	≤4
Phosphate	5.26	5	86.23	0.006	0.25	0.28	≤0.4
Nitrate	187.33	5	93.02	0.266	0.70	4.2	≤50

per gram of material (qeq), 0.648 mg of ammonia, 0.006 mg of phosphate and 0.266 mg of phosphate were removed. High percentage removal overall was observed, with total removal efficiencies of ≥85% being achieved. This system has the potential to be a novel cost effective treatment method for LFL. Further research is ongoing to develop and optimise a large scale on-site treatment system using larger volumes and different concentrations of leachate

4. Conclusion

These results demonstrate that leachate can be treated effectively by bioremediation and adsorption-based systems. LFL represents a major problem for MSW landfills, however, this treatment system may be an effective treatment option within Ireland. The main finding of this study are;

1. Bioremediation is successful at treating LFL, specifically ammonia which achieved a removal efficiency of ≥89%.
2. Adsorption, using low cost material, such as pumice and oyster shells both have the ability to reduce ammonia, phosphate and nitrate levels in LFL
3. As a whole the system employed in the current study effectively achieved discharge limits for the tested compounds, ammonia, phosphate, and nitrate, by 98%, 86% and 93%, respectively.
4. Further research is now required to determine the potential of using this system on larger volumes of LFL which vary in composition and concentration

References

Aksu, Z. and Gönen, F. (2004) 'Biosorption of phenol by immobilized activated sludge in a continuous packed bed : prediction of breakthrough curves', 39, pp. 599–613. doi: 10.1016/S0032-9592(03)00132-8.

Brennan, R. B., Clifford, E., Devroedt, C., Morrison, L. and Healy, M. G. (2017) 'Treatment of land fi ll leachate in municipal wastewater treatment plants and impacts on ef fl uent ammonium concentrations', *Journal of Environmental Management*. Elsevier Ltd, 188, pp. 64–72. doi: 10.1016/j.jenvman.2016.11.055.

Brennan, R. B., Healy, M. G., Morrison, L., Hynes, S., Norton, D. and Clifford, E. (2016) 'Management of landfill leachate: The legacy of European Union Directives'. doi: 10.1016/j.wasman.2015.10.010.

EPA (2015) *EPA Licensed Sites Report on Waste Enforcement*. Available at: https://www.epa.ie/pubs/reports/enforcement/Report_On_Waste_Enforcement.pdf.

Kamaruddin, M. A. (2015) 'Sustainable treatment of landfill leachate', *Applied Water Science*. Springer Berlin Heidelberg, pp. 113–126. doi: 10.1007/s13201-014-0177-7.

Kargi, F. and Pamukoglu, M. Y. (2004) 'Adsorbent supplemented biological treatment of pre-treated landfill leachate by fed-batch operation', *Bioresource Technology*, 94(3), pp. 285–291. doi: 10.1016/j.biortech.2004.01.003.

Kaur, K., Mor, S. and Ravindra, K. (2016) 'Removal of chemical oxygen demand from landfill leachate using cow-dung ash as a low-cost adsorbent', *Journal of Colloid and Interface Science*. Elsevier Inc., 469, pp. 338–343. doi: 10.1016/j.jcis.2016.02.025.

Kheradmand, S., Karimi-Jashni, A. and Sartaj, M. (2010) 'Treatment of municipal landfill leachate using a combined anaerobic digester and activated sludge system', *Waste Management*. Elsevier Ltd, 30(6), pp. 1025–1031. doi: 10.1016/j.wasman.2010.01.021.

McCarthy, S., Moriarty, J., O'Riordan, D. and O'Leary, G. (2010) *Focus on Landfilling in Ireland*, *Environmental Protection*.

McCoole, M. F., Kurz, D. I., McDonagh, M. M., Derham, D. J. and O'Neill, M. D. (2010) *National Waste Report 2010*. Available at: http://www.scew.gov.au/system/files/resources/020c2577-eac9-0494-493c-d1ce2b4442e5/files/wastemgt-nat-waste-report-final-20-fullreport-201005-0.pdf.

Paskuliakova, A., Tonry, S. and Touzet, N. (2016) 'Phycoremediation of landfill leachate with chlorophytes: Phosphate a limiting factor on ammonia nitrogen removal', *Water Research*. Elsevier Ltd, 99, pp. 180–187. doi: 10.1016/j.watres.2016.04.029.

Raghab, S. M., Abd El Meguid, A. M. and Hegazi, H. A. (2013) ‘Treatment of leachate from municipal solid waste landfill’, *HBRC Journal*. Housing and Building National Research Center, 9(2), pp. 187–192. doi: 10.1016/j.hbrcj.2013.05.007.

Rao, J. R. and Viraraghavan, T. (2002) ‘Biosorption of phenol from an aqueous solution by Aspergillus niger biomass’, 85, pp. 165–171.

Renou, S., Givaudan, J. G., Poulain, S., Dirassouyan, F. and Moulin, P. (2008) ‘Landfill leachate treatment: Review and opportunity’, *Journal of Hazardous Materials*, 150(3), pp. 468–493. doi: 10.1016/j.jhazmat.2007.09.077.

Zayen, A., Schories, G. and Sayadi, S. (2016) ‘Incorporation of an anaerobic digestion step in a multistage treatment system for sanitary landfill leachate’, *Waste Management*. Elsevier Ltd, 53, pp. 32–39. doi: 10.1016/j.wasman.2016.04.030.

Zhang, L., Li, A., Lu, Y., Yan, L., Zhong, S. and Deng, C. (2009) ‘Characterization and removal of dissolved organic matter (DOM) from landfill leachate rejected by nanofiltration’, *Waste Management*. Elsevier Ltd, 29(3), pp. 1035–1040. doi: 10.1016/j.wasman.2008.08.020.