Treatability study of two hybrid-passive treatment systems for landfill leachate operated at cold temperature
Sean Speer, Pascale Champagne and Bruce Anderson

ABSTRACT
Cold ambient temperatures can negatively affect the performance of passive and semi-passive landfill leachate treatment systems and decrease treatment efficiency. Cold temperature leachate treatment efficiencies were compared between a commercially available semi-passive treatment system and a passive peat and wood shaving biological trickling filter. The addition of an active fixed-film pretreatment stage in the treatment train was also assessed. Results indicated that the internal temperature of the peat filters was independent of influent water temperature; exothermic reactions maintained internal system temperatures. It was determined that pretreatment of the leachate did not affect the overall removal of chemical oxygen demand (COD), but did increase nitrification in the subsequent passive treatment systems and allowed for the removal of dissolved inorganic constituents prior to the passive treatment system, which will extend the useful life of the entire treatment train. The hybrid-passive treatment systems reduced COD concentrations by 10 ± 3% and 15 ± 3%, in the semi-passive treatment system and the peat and wood shaving biological trickling filter-based systems, respectively, and indicated that nitrifying biomass was starting to populate the treatment systems. It was therefore concluded that operation of these systems would be feasible under cold climate and should be assessed at the pilot-scale.

Key words | cold climate, landfill leachate, passive treatment, pretreatment, sub-surface flow

INTRODUCTION
The cost associated with landfill leachate treatment is one of the largest long-term costs at sanitary landfills and one that continues long after the landfill ceases accepting waste and generating a profit. Leachate is generated as moisture percolates through the stored waste, accumulating by-products from the biochemical degradation of the waste. A number of studies have demonstrated that the strength and volume of the leachate produced is both site specific and variable over the life of a landfill (Maehlum 1995; Bulc 2006; Kinsley et al. 2007; Spagni et al. 2008; Vymazal & Kropfelova 2009). This site-specific nature of landfill leachate requires a site-specific remediation strategy, a factor that is most relevant when designing passive treatment systems.

Leachate treatment strategies can be separated into two main categories: active systems, which are strictly controlled and can have numerous chemical and energy inputs (e.g. reverse osmosis, sequencing batch reactors or activated sludge systems); and passive systems, which aim to minimize chemical and energy inputs while still achieving target treatment efficiencies (e.g. constructed wetlands, trickling filters and soil infiltration systems). The lower operating costs of passive treatment systems make them an ideal option for landfill operators (Boller 1997; Rew & Mulamoottil 1999; Chiemchaisri et al. 2009). The lack of strict operational controls within these systems does, however, make them more susceptible to climatic variability and ambient environmental conditions.

Passive treatment systems in cold climates generally require sub-surface flow (SSF) through a porous media, since exposed water will freeze in sub-zero (Celsius) temperatures. SSF systems maintain a layer of unsaturated porous media above the surface of the leachate, thus...
insulating the saturated zone of the treatment system from the cold ambient temperatures. The primary limitation of SSF systems is the limited oxygen transfer efficiency, which can be overcome by decreasing the loading rate to the system, adding supplemental aeration systems, or modifying the flow regime in the system. A number of studies have demonstrated the advantages of vertical sub-surface flow systems (VSSF) over horizontal (HSSF), including the potential for larger loading rates, better oxygen transfer and higher oxidizing conditions (Pendleton et al. 2005; Kruzic & Liehr 2008; Faulwetter et al. 2009; Forquet et al. 2009; Nivala & Rousseau 2009). Intermittently dosed VSSF systems can achieve the required oxygen transfer via the infiltration of air between dosing events, thus achieving adequate oxygen transfer without the added cost of an aeration system.

Biological conversion of leachate constituents is dependent on both contact time and temperature. Lower operating temperatures decrease the activity of the biological consortia within a treatment system. Lower biological and chemical reaction rates require longer contact times between the microorganisms and the leachate, thus requiring larger treatment system sizes. SSF treatment systems can maintain adequate temperatures for treatment if they are sufficiently insulated from cold ambient temperatures, since the microbial activity in the treatment cells tends to be exothermic. With adequate insulation Nivala et al. (2007) noted significant treatment even with ambient air temperatures below −20 °C.

An active pretreatment system can be combined with a passive treatment system producing a hybrid-passive technology. The active pretreatment reduces the organic loading, thereby reducing the oxygen demand experienced by the passive treatment system. This type of treatment strategy can minimize the size of both the active and the passive treatment stages, allowing for cost optimization. The pretreatment stage of the hybrid-passive treatment system can also serve to accommodate sedimentation, allowing for the removal of inorganic solids that could clog the pore spaces of the subsequent packed bed passive treatment system. Rustige & Nolde (2007) monitored a passive treatment wetland with and without pretreatment and found that the pretreatment system had no effect on final effluent quality; however the precipitation and settling of inorganic constituents within the wetland cells (and the decrease in hydraulic conductivity associated with this precipitation) was lower in the wetland receiving pretreated leachate. It was concluded that the pretreatment system removed a substantive fraction of the inorganic leachate constituents that could adversely affect the flow in the wetland cells without pretreatment of the leachate.

This bench-scale treatability study evaluated the effectiveness of two treatment systems in a hybrid-passive treatment system for potential implementation at the Merrick Landfill, which serves the City of North Bay, Ontario, Canada. The landfill is located in the Near North region of the Province of Ontario where the average ambient air temperature fluctuates between 25 and −18 °C. This bench-scale study also investigated the effect of pretreatment on the removal of leachate constituents in the subsequent passive treatment systems under cold ambient temperatures.

METHODS

The bench-scale treatability study was conducted in a temperature-controlled chamber (2 ± 2 °C). The purpose of the study was to analyse the efficiencies of two different landfill leachate treatment systems, a passive peat and wood shaving biological trickling (PW) filter designed by Queen’s University and a commercially available semi-passive treatment (SP) system exposed to cold ambient temperatures. Two of each of the treatment systems were constructed; one system was dosed directly with raw leachate, while the other system was dosed with pretreated leachate. All of the systems were previously investigated at the bench-scale at room temperature, and the loading rates for this study were selected based on the findings from this previous bench-scale study (Speer et al. 2010). The landfill leachate used in this study was supplied and shipped directly from the Merrick Landfill in North Bay, Ontario, Canada.

Pretreatment system

Pretreatment of the leachate was performed using a fixed-film aerobic pretreatment reactor with a 3-day hydraulic residence time as described by Speer et al. (2010). A single pretreatment system was used in this study to ensure that...
each of the treatment systems was dosed with the same influent stream. The pretreatment system was a circular high-density polyethylene reactor with diameter of 30 cm and a liquid depth of 23 cm. The reactor was packed with an inert plastic media (1% of total reactor volume) to promote biological attachment.

**Semi-passive treatment system**

The SP systems (Figure 1(a)) consisted of two 1.2 m (4 ft) long, 10 cm (4 in) I.D. PVC pipes in series, packed with a proprietary medium. Loading rates to the systems were set at 0.41 m$^3$/m$^2$/d, with raw leachate dosed to SP1 and pretreated leachate dosed to SP2. Full-scale versions of these treatment systems are equipped with fans to enhance air movement within the cells (for aeration); this was replicated in this bench-scale study through the addition of pressurized air to the SP systems. Since the SP systems were actively aerated, they were therefore dosed with leachate continuously as opposed to employing a cycle of dose-and-rest to achieve passive aeration.

**Peat and wood shaving biological trickling filter**

The two bench-scale PW filters were constructed using 36 cm (16 in) tall, 10 cm (4 in) I.D. Plexiglas columns (Figure 1(b)). The outlet of the columns consisted of a standpipe that maintained the lower 20 cm (8 in) of the columns saturated for biological treatment of the leachate. The treatment systems were dosed at a loading rate of 0.31 m$^3$/m$^2$/d, with raw leachate dosed to PW1 and pretreated leachate dosed to PW2. These biofilters were packed with a mixture of 25% peat and 75% wood shavings (v/v) while water was recirculated through the columns, allowing the media to settle to a natural saturated density (240 kg/m$^3$). These systems were passively aerated by applying leachate to the filter in 6 doses of 1 h (0.4 L per dose), followed by a rest period of three hours during which the leachate could percolate through the media.

**System setup**

The Merrick landfill intends to install a co-generation system to combust the landfill gas collected on site to produce energy and reduce greenhouse gas emissions. The waste heat from the co-generation system could be transferred to the raw leachate entering the treatment system, increasing influent temperatures and biological activity in the system during cold weather operations. This was simulated in the bench-scale laboratory study by maintaining the raw leachate at approximately 24°C, while the treatment systems, including the pretreatment system, were operated in the temperature-controlled chamber at 2°C. It should be noted that a temperature difference in the influent existed between
the systems under investigation. The pretreatment system and the treatment systems dosed directly with raw leachate (SP1 and PW1) received influent leachate at approximately 24 °C, while the treatment systems dosed with pretreated leachate (SP2 and PW2) received pretreated leachate at approximately 5 °C. All SP systems and PW filters were insulated using 3.8 cm (1.5 in) duct wrap insulation to simulate the natural insulation provided in a buried system; 2.5 cm (1 in) of gravel and 10 cm (4 in) of peat and wood shaving mixture were added above the leachate dosing pipe of the PW filters for insulation. All of the hybrid-passive treatment system components were previously used for a room-temperature bench-scale study (Speer et al. 2010) and the biological consortia in the systems was assumed to be viable, therefore the systems were not seeded prior to initiation of the cold-temperature study.

Sample and data analysis

As the systems were dosed with small leachate volumes, composite samples were collected over a 12-h period to produce sufficient sample volume for analysis. Samples were collected and analysed weekly in accordance with Standard Methods for water quality (APHA 1998). Ammonia (4500-NH3 D.), nitrate-nitrite (4500-NO3 B.), chemical oxygen demand (COD) (5220 D.), standard alkalinity (320 B.), and total Kjeldahl nitrogen (TKN) (4500-Norg B.) were measured using ion chromatography. Nitrite and nitrate concentrations were measured using an inductively coupled plasma atomic (optical) emissions spectrometer. Iron, calcium and manganese concentrations were measured using ion chromatography.

Treatment efficiencies of the leachate constituents were evaluated on a cumulative mass removal basis for all of the treatment system configurations. Cumulative mass flux was employed to dampen the effects of influent leachate variability and minimize the potential effects of retardation on the various leachate constituents within the treatment system cells due to adsorption-desorption or impingement-sluoughing processes that could affect the effluent concentrations. The cumulative mass removals were calculated using Equation (1):

\[ \%R^{n} = \frac{\sum_{i=1}^{n} (Q_{\text{inf}}^{i} C_{\text{inf}}^{i} - Q_{\text{eff}}^{i} C_{\text{eff}}^{i}) \times 100}{\sum_{i=1}^{n} Q_{\text{inf}}^{i} C_{\text{inf}}^{i}} \] (1)

where \( \%R \) is the cumulative percentage mass removal [%], \( Q \) is the flow rate [L/T], and \( C \) is the concentration [M/T]. In the equations, the superscript \( (n) \) represents the sampling event, and the subscripts represent the location of the samples (the influent or effluent from the treatment system stages).

JMP, a statistical data processing program, was used to compare the data from the various treatment system setups. Wilcoxon rank sum tests were applied to determine if differences in the influent and effluent concentration data from each treatment system were statistically significant. The statistical significance from the tests was based on a 95% confidence interval \( (p < 0.05) \).

RESULTS AND DISCUSSION

The raw leachate used in this study was characterized as having a circumneutral pH and high alkalinity (4,000–4,500 mg/L as CaCO₃), which decreased slightly throughout the duration of the study. The COD concentration of the raw leachate varied between 1,000 and 1,500 mg/L with a decreasing trend (Figure 2) that was similar to the trend noted for ammonia, which varied between 550 and 450 mg/L (Figure 3) between the onset and completion of the bench-scale study.

The pH of the raw leachate (Figure 4) remained relatively constant \( (7.2 \pm 0.1) \) with the exception of one spike in values to approximately 8. A study conducted at the University of Western Ontario (2010) reported that the labile organic matter in the raw leachate from the Merrick Landfill comprised 20% volatile fatty acids. These acids were removed from the leachate throughout the various treatment system stages via aerobic, biologically mediated degradation, as well as abiotic processes such as volatilization. Removal of these acids would reduce the acidity of the leachate thereby increasing the pH, as has been reported in other studies involving passive and semi-passive treatment systems (e.g. Jowett et al. 1999; Martensson et al. 2007). Figure 4(b) shows that pretreatment of the leachate affected the effluent pH from the SP systems \( (p = 0.01) \) and the PW filters \( (p < 0.01) \). The effluent from SP2 (dosed with pretreated leachate) maintained pH levels similar to, but slightly higher than, those measured in the pretreatment system \( (8.6 \pm 0.1) \) in the pretreatment system, \( (8.8 \pm 0.1) \) in SP2, \( p < 0.01 \). The effluent pH from SP1 (dosed with raw leachate) was initially in the same range as SP2 \( (8.8 \pm 0.0) \), and decreased throughout the duration of the study to approximately \( 8.3 \pm 0.1 \) (Figure 4).
The PW filters maintained lower overall pH values than the SP systems, likely because of the humic and fulvic acids that are regularly leached from peat (Champagne et al. 2005).

The cold ambient temperatures were expected to reduce biological activity; however there were still notable removals of both COD and total nitrogen concentrations. COD concentrations in the raw leachate decreased from 1,200–1,500 mg/L initially to concentrations ranging from 800 to 1,000 mg/L in weeks 5 through 8 of the study (Figure 2). Removal of COD in the systems treating raw leachate and those treating pretreated leachate was equivalent ($p = 0.37$ and $p = 0.85$, respectively, in the SP systems and

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**Figure 2** | (a) Weekly COD concentrations; (b) means and standard deviations of COD concentrations. SP1 and PW1 dosed with raw leachate, SP2 and PW2 dosed with effluent from the pretreatment system. SP denotes the semi-passive treatment systems, and PW denotes the peat and wood shaving biological trickling filters.

**Figure 3** | (a) Ammonia, (b) nitrite, (c) nitrate, and (d) total nitrogen concentrations reported as mg-N/L. SP1 and PW1 dosed with raw leachate, SP2 and PW2 dosed with effluent from the pretreatment system. SP denotes the semi-passive treatment systems, and PW denotes the peat and wood shaving biological trickling filters. See Figure 2 for explanation of abbreviations.

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the PW filters), and therefore the inclusion of a pretreatment stage in the leachate treatment system showed no measurable effect on the overall removal of COD from the treatment systems. However, it was evident that the pretreatment system provided some COD removal (10 ± 5%), which led to a lower loading of COD to the subsequent treatment systems. This decrease in loading would lead to a decrease in the biological activity required for COD reduction and, consequently, a reduction in the volume of biomass in the treatment systems. The extent of biomass-mediated clogging in the treatment system pore space would, therefore, also be reduced with the addition of the pretreatment system.

The addition of the pretreatment stage in the leachate treatment system had the largest influence on overall ammonia removal (Figure 3(a)), but only with respect to the PW filters. Both of the SP systems removed ammonia (36 ± 4% and 36 ± 6% in SP1 and SP2, respectively); however the removals were not determined to be statistically different (p = 0.33). The ammonia removal in the PW filters was highly dependent on the addition of the pretreatment stage. The raw leachate-dosed PW1 experienced no significant removal of ammonia (5 ± 2%), while the pretreated leachate-dosed PW2 removed 25 ± 2% of the influent ammonia (after the onset of nitrification). It was therefore evident that the addition of the pretreatment system increased the overall ammonia removal in the PW filters.

Conventional nitrification involves the biologically mediated conversion of ammonia to nitrite (nitrification), followed by further oxidation of nitrite to nitrate (nitratation). This biologically mediated conversion was only observed in the effluent of PW2, which was characterized by an initial increase in nitrite concentrations (Figure 3(b)), followed by a decrease in nitrite concentrations correlated with an increase in nitrate concentrations (Figure 3(c)). From this, it was concluded that the ammonia removal mechanism in this treatment system was likely conventional nitrification. Nitrite was also detected in the effluent from PW1 at various times throughout the study (Figure 3(b)). These increases in nitrite concentrations were not associated with an increase in nitrate concentrations (Figure 3(c)), which would have indicated complete conventional nitrification, and the nitrite concentrations decreased again at the end of the study. Since conversion of ammonia to nitrite was not consistent in PW1, it was, therefore, assumed that volatilization and biomass assimilation were the likely ammonia removal mechanisms in this system.

Both of the SP systems exhibited statistically significant removals of total nitrogen (p = 0.02 and p < 0.01 in SP1 and SP2, respectively) without the formation of nitrite or nitrate (Figure 3). PW2 also exhibited a reduction in total nitrogen (Figure 3(d)) for the first 4 weeks of the study (p = 0.03), prior to the onset of conventional nitrification. The average pH of the pretreated leachate dosed to the treatment systems was 8.7 ± 0.1, and the effluent pH of the treatment systems were 8.6 ± 0.2, 8.1 ± 0.1 and 8.2 ± 0.1 in SP1, SP2 and PW2, respectively. These pH values were lower than those suggested for air-stripping systems designed for ammonia removal (Metcalf & Eddy 2003). The pH and temperature values were, however, in a range such that approximately
10–20% of the ammonia present in the leachate would be in the unionized form that could be removed through volatilization. The percentage removals of nitrogen in SP1 (36 ± 4%), SP2 (39 ± 8%) and PW2 (15 ± 8% prior to onset of nitrification) supported the assumption that volatilization was the likely contributing removal mechanism.

In both types of treatment system, flow and treatment of leachate was designed to occur in the pore spaces of the packed beds. Clogging of these pore spaces is a concern and a specific design consideration, especially in treating landfill leachate containing high concentrations of dissolved inorganic compounds. Jowett et al. (1999) reported on the use of a Waterloo Biofilter system at a landfill site, stating that within 2 years of commissioning the landfill leachate treatment system, the flow distribution system became clogged with carbonate minerals, and after 4 years a crust of iron precipitates had formed on the top layer of the biofilter media. One of the main design goals of the pretreatment system used in this study was to minimize clogging of the media through the induced oxidation and precipitation of dissolved inorganic constituents.

Comparing the concentrations of iron, calcium and manganese (Figure 5), it was evident that the pretreatment system allowed for the precipitation and removal of these cations (89 ± 2%, 87 ± 4% and 83 ± 2% for calcium, iron and manganese, respectively), which reduced the potential for their precipitation in the subsequent treatment systems. Iron precipitation from landfill leachate results from the oxidation of reduced ferrous iron (the dominant form in the leachate) to ferric iron and the subsequent precipitation as ferric hydroxide (Nivala et al. 2007). Therefore the precipitation of iron required oxidizing conditions, which were present in all of the treatment systems, and as such all of the treatment systems reduced iron concentrations to approximately the detection limit of the analytical method (2 mg/L) independent of raw or pretreated leachate dosing. The pH of the raw leachate (7.2 ± 0.1) indicated that bicarbonate (HCO$_3^-$) was likely the dominant form of inorganic carbon in the leachate. Once the pH increases above 8.3, carbonate (CO$_3^{2-}$) may become more significant (Snoeyink & Jenkins 1980). The pretreatment system pH was 8.6 ± 0.1 indicating that carbonate would be available for precipitation of calcium carbonate (CaCO$_3$) and manganese carbonate (MnCO$_3$).

It could be argued that the addition of the pretreatment system exhibited negligible effects on the overall removal of iron, calcium and manganese (Figure 5). The addition of the pretreatment system did, however, allow for the precipitation of dissolved inorganic constituents prior to dosing of the treatment systems. This would decrease the potential for, and magnitude of, precipitation of inorganic constituents in the pore spaces of the treatment systems, increasing the operational lifespan of the system. The effluent dissolved calcium and manganese concentrations from the systems dosed with pretreated leachate were not statistically different from the effluent concentrations from the pretreatment system (0.55 < p < 1 for Ca, and 0.13 < p < 0.17 for Mn). This indicated that the addition of the pretreatment stage in the treatment systems would reduce the removal of dissolved inorganic constituents in the subsequent treatment systems, and consequently reduce the potential for clogging.

The space-constrained layout of the experimental system on this scale presented limitations affecting performance. It was expected that both types of treatment system would effectively reduce leachate strength even at cold temperatures, as has been noted in other studies (e.g. Jowett et al. 1999; Nivala et al. 2007). At the end of the study, the internal temperature of each of the systems was measured. It was noted that the PW filters maintained internal temperatures of 11 °C regardless of the differences in influent leachate temperature (24 °C for the raw leachate and 5 °C for the pretreated leachate). This indicated that heat was generated by exothermic biologically mediated reactions within each system. This was an important finding that indicated that larger field-scale systems, with more volume and biological activity, could feasibly maintain adequate internal temperatures to promote efficient biological activity independent of the cold ambient temperatures or cold influent leachate temperatures. The SP systems were aerated with cold air, which led to internal temperatures equal to the ambient temperature (2 °C). In field applications, the SP systems utilize internal fans to move air, rather than supplying air through pumps. As such, the biologically mediated exothermic reactions that were responsible for maintaining the internal temperature of the PW filters would also be expected to maintain adequate internal temperatures in the SP systems. The low internal temperatures of the SP systems in this study were therefore a result of the experimental protocol rather than an indication of full-scale system design performance.
CONCLUSIONS

The pretreatment system used in this study demonstrated that, even at cold temperatures, removal of COD could be achieved. This COD removal in the pretreatment system allowed for more nitrification to occur in the PW filter treating pretreated leachate, than in the PW filter treating raw leachate. The pretreatment system also allowed for the precipitation of dissolved inorganic constituents prior to dosing of the packed bed treatment systems, which reduced the potential for inorganic precipitates to clog the downstream treatment systems. This would extend the useful life of these systems, as well as reduce operational and maintenance costs.

Although insulation of the systems would be more complete for these systems when implemented on a larger scale, this study did, however, demonstrate that the temperature in each of the PW filters was independent of influent temperature. Even at this small scale and with limited insulation, temperatures were maintained at 11°C within the PW filters while the ambient temperature was 2°C. Air movement in the bench-scale SP systems was a limiting factor in maintaining the internal temperatures; full-scale application of these systems would likely require more complete insulation to maintain consistent temperatures throughout.
systems would minimize the introduction of cold ambient air into the system and would overcome this limitation. Hence it was concluded that if these systems were designed and implemented on a larger scale and provided with more insulation as would be expected in the field, the internal temperatures could be maintained at levels that would sustain biological activity even under cold ambient temperature conditions, as well as with cold temperature influents.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the support of SNC Lavalin Inc., and the City of North Bay, as well as the financial support of the Ontario Centres for Excellence.

REFERENCES


