Influence of landfill leachate suspended solids on clog (biorock) formation

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Abstract

Laboratory column tests were performed to evaluate the role of leachate-suspended-solids in clogging a granular material permeated with Keele Valley Landfill leachate. The development of the clog material was a result of biological, chemical, and physical processes occurring within the column. The increase in volatile solids, which contributed to clog development over time, was primarily due to the retention of volatile suspended solids and growth of a biofilm capable of removing acetate, propionate, and butyrate from the leachate. Acetate fermentation was primarily responsible for precipitation of calcium within the column. The precipitated calcium and retention of inorganic suspended solids contributed to the increase in clog inorganic solids. Over the duration of the experiment, 3.7 times more calcium was precipitated in the column (due to acid fermentation) than was retained with inorganic suspended solids. Clogging resulted in a greater than 60% reduction in drainable porosity and a six-order magnitude decrease in hydraulic conductivity. The potential practical implications with respect to pipe cleaning and leachate recirculation were discussed.

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1. Introduction

Leachate collection systems (LCS) are used at the base of most modern landfills to limit the advective flux of contaminants through the barrier system, hence minimizing the potential for groundwater contamination below the landfill. LCS typically consist of perforated HDPE collection pipes embedded within a granular drainage blanket that is positioned below the waste and above any low permeability liners. The clogging of these systems due to a build-up of what is sometimes called “biorock” is of increasing concern to landfill operators. Field studies of numerous German landfills by Brune et al. (1991) demonstrated that the drainage material in collection systems can clog due to biological, and chemical, and physical processes. It was suggested that the clog material formed as a result of a high population of microorganisms that accelerated the precipitation of minerals out of leachate, leading to a reduction in pore volume of the media. The chemical composition of the clog material (dry mass) in the German study averaged about 21% calcium, 34% carbonate, 16% silica, 8% iron, and 1% magnesium. Fleming et al. (1999) reported organic and inorganic clogging in the saturated component of a collection system that was exposed to leachate for about four years at the Keele Valley Landfill (KVL) in Maple, Ontario. Chemical composition of this clog material was similar to that of the German study with 20% calcium, 30% carbonate, 21% silica, 2% iron, and 5% magnesium on a dry mass basis. Samples removed from the system ranged from soft clog material containing solid sand-size particles to hard solid materials with the appearance and consistency of a weak concrete or rock.

Rowe et al. (2002) reported the results of laboratory column experiments where a gravel-size medium was permeated with synthetic leachate. The synthetic leachate approximated the average chemical composition of

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KVL leachate in the early life of the landfill, but contained relatively low suspended solids and biological activity compared to real (KVL) leachate. These experiments provided insight regarding the role of acetate fermentation in precipitating calcium carbonate \((\text{CaCO}_3)\) from leachate and assessed clogging due to biomass growth and \(\text{CaCO}_3\) accumulation with time along the leachate flow path. Rowe et al. (2002) reported the rate of clogging was greater in a column operated with KVL leachate relative to a synthetic leachate due, in part, to the accumulation of suspended solids from KVL leachate within the pore space of the gravel-size material. Armstrong (1998) and Rowe et al. (2000a,b) completed similar column experiments as Rowe et al. (2002) that operated with KVL leachate to examine the effects of temperature, mass loading (in terms of flow rate), and particle size on the rate of clogging. These studies showed that, as leachate passed through the gravel-size medium, there was a significant reduction in chemical oxygen demand (COD) and precipitation of calcium \((\text{Ca}^{2+})\). However, the role of the suspended solids with respect to the rate of removal of volatile fatty acids (VFA) with position in the column was not established. Additionally, the column studies by Armstrong (1998) and Rowe et al. (2000a,b) were directed at assessing clog material properties after significant clogging occurred; thus, these studies did not provide insight regarding the distribution and composition of clog at times prior to significant clogging.

The objective of this study was to assess the role of suspended solids on the formation of active biofilms and accumulation of clog solids within the pore space of a gravel-size medium permeated with KVL leachate. Particular attention is paid to the changes (if any) in clog composition after different periods of leachate treatment and to the amount of clogging attributable to biological growth, precipitation of minerals, and filtration/straining of suspended particles. The data was collected with the objective of gaining insight regarding how the various clogging mechanisms change with time and to provide experimental data that could be used to calibrate geochemical models of clogging (e.g., Cooke et al., 1997; Cooke et al., 2001). This understanding is also important for assessing practical means of dealing with clogging in LCS. In contrast to previous studies, the variation in clog composition and the consumption of VFAs in real (KVL) leachate with position and time is examined.

2. Methodology

Four identical columns packed with 6 mm diameter glass beads were permeated under anaerobic conditions with leachate (from the bottom) at a design flow rate of 0.51 m³/m²/day at 21 °C. These are the same conditions used in the column study reported by Rowe et al. (2002), except for the replacement of synthetic leachate with KVL leachate.

Each column consisted of 50.8 mm diameter, 700 mm long, PVC monitoring well pipe (Schedule 40) with a flush joint cap and plug, and mounted in an upright position. Each column was equipped with an influent near the column base and an effluent valve just above the top of the gravel-size medium (beads) (Fig. 1). The medium length was 360 mm. Piezometers and sample ports were positioned at 60 mm intervals above the column base screen to a height just below the effluent valve. Each piezometer consisted of a nylon elbow barbed fitting tapped into the column and fitted with clear nylon tubing to allow for the monitoring of the leachate head, and the removal of any gas generated within the columns. The piezometers were located closer together in this study than in previous studies so as to provide additional detail regarding the reduction in hydraulic conductivity of the granular medium as it clogged. Each column was separated into six sections. A section is defined as the length of the medium between two adjacent piezometer locations, with the first section \((\text{S1})\) starting above the base screen, and the last section \((\text{S6})\) finishing at the top of the column (Fig. 1). On the side opposite each piezometer, a leachate sample port was tapped into the column.

Leachate samples were collected from the sample ports along the column and tested for COD, Ca²⁺, acetate (AA), propionate (PA), and butyrate (BA) concentrations on a weekly basis. To provide discrete position concentrations of measured parameters, 1 to 3-ml of leachate was removed from each sample port. To complete the desired testing program, leachate influent and effluent were tested for pH, total (TSS) and volatile (VSS) suspended solids.

COD concentration was measured using a Hach™ COD reactor with Hach™ COD reagents that heated the reagent and leachate at 150 °C for 2 h followed by an analysis with a Hach™ DR2000 Spectrophotometer. Ca²⁺ concentrations were obtained using a Philips PU9100X atomic absorption spectrometer. TSS and VSS were tested using a gravimetric measurement of the residue retained on a 0.45 μm glass fiber filter dried at 105 and 550 °C, respectively. VFA concentrations were obtained by gas chromatography (GC), using a Shimadzu GC-9A and Varian 3400 instruments equipped with flame ionization detectors and 15 m × 0.53 mm (or 0.25 mm) inner diameter, 0.5 μm film NUKOL™ (Supelco, Bellfonte, PA) capillary columns. Injections to the Varian 3400 were automated (Varian 8200 auto sampler) and performed using solid phase microphase extraction (SPME) with 10-min immersion/2-min desorption of a 75 m Carboxen™/Polydimethylsiloxane fiber (Supelco, Bellfonte, PA). All aqueous VFA samples were acidified in 1% H₃PO₄ prior to analytical
separation. Isovaleric acid was used as an internal calibration standard.

Drainable porosities were calculated twice a month to quantitatively assess the amount of clogging that occurred within the columns with time. The drainable porosity was calculated by draining a section of the column, measuring the volume of fluid collected, and dividing this by the total volume within that section, while correcting for fluid removed from piezometers. Due to surface tension and fluid trapped in void spaces and biofilm, some fluid could not be drained from the column. Therefore, the drainable porosity represents the pore space that will freely drain fluid in a gravitational field under atmospheric pressure. In early stages of clog development, the drainable porosity provides a useful indication of the degree of clogging. In the late stages of clogging, the drainable porosity drops to zero, but this does not imply zero actual porosity (as described above).

The initial hydraulic conductivity, based on a modified version of ASTM (2000) (D2434-68), of the 6 mm diameter glass beads was 0.33 m/s. This value was used as a benchmark to assess how the hydraulic conductivity decreased with time and drainable porosity. After initiation of the experiments, the hydraulic conductivity was only obtained once there had been sufficient clogging to give a measurable difference in leachate level between adjacent piezometers. Once this head difference was known over a section of the column, the hydraulic conductivity was calculated from the known flow length and flow rate. At a flow rate of about 0.51 m³/m²/day (selected to be typical of flow in a leachate collection system near drainage pipes, see Rowe et al., 2000a), a measurable head difference between adjacent piezometers did not occur until the hydraulic conductivity dropped by about four orders of magnitude.

The columns were disassembled after four periods of leachate COD removal (described below) to allow an assessment of the changes (if any) in clog composition with time. At disassembly, the columns were cut open, and the beads and clog material were removed from specific zones in the column (0–100, 100–200, 200–300, and 300–360 mm). The beads and clog material were oven dried (105 °C) to obtain a water content. They were then placed in a furnace (550 °C), and the organic and inorganic contents were calculated. The ash (non-combustible solids, denoted as NVS) density of the clog material remaining after volatilization at 550 °C for 15 min was obtained using ASTM (2002) (D854).

3. Influent and effluent leachate composition changes

Due to the natural variability of real landfill leachate, the influent characteristics varied on a daily and weekly basis. The average influent COD and Ca²⁺ concentrations were 12,200 and 460 mg/l, respectively, with an average pH of 6.8 (Fig. 2). The average influent AA, PA, and BA concentrations were 3340, 3940, and 1610 mg COD/l, respectively, giving an average total calculated...
The COD concentration of 8890 mg COD/l (Fig. 3). Thus, the three measured volatile acids comprised about 73% of the total COD, indicating that other, more complex organics were also present in the KVL leachate. The average influent TSS and VSS concentrations were 800 and 350 mg/l, respectively, each with a relatively high average standard deviation of 700 and 300 mg/l, respectively.

The changes in AA, PA, and BA concentrations between the influent and effluent of the column are shown in Fig. 3. During the first 70 days: (1) the effluent AA concentration was higher than the influent, (2) influent and effluent PA concentrations were similar, and (3) the effluent BA concentration significantly decreased. The decrease in BA likely caused the increase in AA, because AA is produced by the fermentation of BA (Bjerg et al., 1995; Parkin and Owen, 1986). The lag time for BA removal to occur was within the first 15 days of column operation. Since AA is produced during BA fermentation, it is difficult to assess the lag time for AA removal; however, the lag time likely occurred between 15 and 50 days. After 70 days, AA and BA effluent concentrations were lower than the influent concentrations. PA experienced some removal (about 20%) after about 150 days. The longer period of time for PA to degrade, compared to AA and BA, is likely attributed to the slower growth rate of bacteria that oxidize PA to AA (Boone and Bryant, 1980). Since the three measured volatile acids comprised about 73% of the total COD, it is possible that the remaining 27% of the COD could experience some removal within the column. Comparing the removal of total calculated COD attributed to AA, PA, and BA removal with the total COD removed, it appears that about 500–1000 mg COD/l of non-AA, PA, and BA organics degraded within the column, with most of this removal occurring within the lower half (i.e., near the influent end) of the column.

The influent pH varied between 6 and 7 during the testing program (Fig. 2). The effluent pH was marginally higher than the influent pH (ranged 6.4–7.4) during the first 50 days of operation. After 150 days of operation, there was a gradual increase in effluent pH from near neutral to a value of 7.4–7.5. Similar effluent pH values were obtained from column studies performed using
synthetic leachate (Rowe et al., 2002). In order to assess the clog material properties with time, columns were disassembled at different points in time. Clog chemical composition analysis included inductively coupled plasma spectrometry (ICP) and inductively coupled plasma mass spectrometry (ICP/MS) for major and trace elements, instrumental neutron activation analysis (INAA) for Co and Cr, pulse gamma neutron activation (PGNAA) for B, and LECO method for total and organic carbon, S, SO₄, and CO₂. VK-1 was disassembled at 57 days, after 29 m³/m² of leachate had passed and there was about 10% removal of COD within the column. VK-2 was disassembled at 115 days (53 m³/m² of leachate passed) and VK-3 was disassembled at 157 days (74 m³/m² of passed leachate), and there was about 30% removal of COD within the columns at these times. The last column, VK-4, was disassembled at 157 days, after 114 m³/m² of passed leachate and COD removal of about 50%. To be consistent with previous column studies, the time required to obtain 10% removal of COD is called the lag phase. The lag phase was about 57 days and was followed by a time when there was a large change in COD removal, called the transition phase. The transition phase was followed by a steady state phase starting at about 115 days. As discussed below, each VFA experienced a different lag, transition, and steady state phase. Thus, unless specified otherwise, the lag, transition, and steady state leachate treatment periods refer to a bulk measure of the organic acid removal as COD.

4. Clog development

4.1. Lag phase

It is hypothesized that microorganisms grow in the buried refuse and are transported into the collection
system where they colonize the drainage stone (Rowe et al., 1995). The leachate used in this study was collected from a holding tank after leachate had entered into and passed through the leachate collection system at the Keele Valley Landfill and contains microorganisms that grew either within the waste or drainage stone. Thus, the suspended solids in KVL leachate contains active suspended biomass. During the first 50–100 days of column operation, there was about a 60% reduction in VSS between the influent and effluent leachate. After this time, about 70–95% VSS removal occurred. Thus, landfill bacteria are being retained within the column, where they may grow as a biofilm.

Fig. 4 shows the concentration of AA, BA, and Ca$^{2+}$ along the length of the column at various elapsed times. At 16 days, there was a small decrease in AA and BA concentration along the column length, and also a small decrease in Ca$^{2+}$ concentration. After 55 days, there was an increase in AA concentration (relative to the influent concentration) moving up the column and this corresponded to a decrease in BA concentration along the column (for each mole of BA fermented, 2 moles of AA are produced: Bjerg et al., 1995; Parkin and Owen, 1986). At this time, there was no significant change in the Ca$^{2+}$ concentration along the column. At 84 days, the AA, BA, and Ca$^{2+}$ concentration all decreased moving up the column (from the influent to effluent end). The decrease was greater in the lower half of the column. There was very little removal of PA during the first 84 days of column operation. Comparing the 16, 55, and 84-day AA, BA, and Ca$^{2+}$ concentration profiles, only minor removals of Ca$^{2+}$ occurred at 16 and 55 days (when AA removal was small) and significant Ca$^{2+}$ removal occurred at 84-days (when significant AA was removed). The data indicates that acetate fermentation (not BA fermentation) primarily drives the Ca$^{2+}$ precipitation. Based on theoretical considerations, Rittmann et al. (1996) concluded that, as leachate passes through the drainage material, depletion of Ca$^{2+}$ would be directly related to the loss of COD due to the fermentation of acetic acid and the consequent generation of carbon dioxide (CO$_2$) and formation of carbonic acid (H$_2$CO$_3$). This results in an increase in pH and carbonate concentration, both of which allow, or accelerate, the chemical precipitation of CaCO$_3$($s$). The present experimental findings are consistent with these predictions.

Fig. 5 shows the mass of total dry solids and volatile (combustible) solids that accumulated within the pore space of the medium at the time of column disassembly. The increase in clog material is a result of the growth of microorganisms, accumulation of mineral solids, and the retention of suspended particles within the porous medium. At 57 days, only a very small mass of volatile solids was retrieved during column disassembly at elevations above 30 mm and zero mass is thus shown in Fig. 5. The large mass of volatile solids within the first segment (compared to all subsequent segments) is consistent with the relatively large removal of BA within the section S1 and the small removal of BA within sections S2–S6 at 55 days (Figs. 4 and 5).

The accumulation of clog material within the column can be described in terms of a volatile and an inorganic film. The volatile film contains (1) the active biomass where microorganisms grow and substrate is utilized and (2) non-active biomass. The development of the volatile film is the result of biofilm development and the retention of volatile suspended solids. The inorganic film consists of precipitate material, retention of inorganic suspended material, and non-degradable biomaterial. If
one assumed two distinct (volatile and inorganic) uniform films form on the bead surface, then the volatile film thickness can be calculated using the method of Rittmann and Brunner (1984), while the inorganic film thickness can be deduced as outlined by Rowe et al. (2002). At 57 days, there was about 10% removal of COD within the column, the calculated volatile film thickness, within 0–30 mm above the base screen, was about 0.120 mm (Table 1). At this time, the inorganic film thickness could not be calculated because it was so thin that there was inadequate inorganic mass of material retrieved from the column to complete a density measurement. A similar but smaller volatile film thickness of 0.09 mm was measured within 0–100 mm above the base screen in a column study that used synthetic leachate instead of KVL leachate (VanGulck, 2003) when there was a 10% removal of COD within the column. The time needed to remove 10% of the COD in the synthetic leachate study was about 250 days (190 days longer than in this study). The synthetic leachate columns were inoculated with typical landfill bacteria using a mixture of KVL leachate and synthetic leachate for nine days. Following this, there was negligible VSS loading and, at the end of the lag phase, only about 100 mg of VSS were retained in the synthetic columns compared to about 9000 mg of VSS in the current tests at the end of the lag phase. Thus, the synthetic leachate columns experienced a negligible mass loading of VSS compared to the KVL leachate columns. The significantly larger mass of VSS retained, and the much shorter lag phase in the KVL columns compared to the synthetic series, suggests that the retention of biomass on the granular medium was dominant over biomass growth in the development of a biofilm, capable of removing about 10% of the influent COD within section S1. The fact that VFA fermentation occurred within section S1 of the column at times much earlier than within sections S2–S6, suggests that a larger mass of VSS was retained within section S1, which is consistent with the data given in Fig. 5.

The TSS in the KVL leachate includes VSS (discussed above) and inert suspended solids (called fixed suspended solids, FSS), where TSS is the sum of VSS and FSS. FSS may be comprised of mineral precipitate and/or soil particles (i.e., silt). The heavier sand particles had already settled out of the leachate (either in the holding tank at the landfill or in the laboratory) before the leachate entered the columns, as suggested by the low silicon content in the clog material retrieved during column disassembly (discussed below). If no suspended particles had settled out of the leachate prior to entering the columns, the rate of clogging would have increased, assuming all other factors are similar. Practical implications with respect to LCS clogging and filter layer design are discussed below. During the first 50–100 days, there was about a 60% removal of FSS between the influent and effluent leachate, and about 70–95% removal after this time – similar to the VSS removal with time.

The retention of suspended solids (FSS and VSS), growth of active biomass on the medium surface, and precipitation of minerals (mainly CaCO\(_3\)\(_s\), discussed below) caused a reduction in the pore space of the medium. The initial drainable porosity of the medium was 0.386 (actual porosity was 0.396), averaged over the entire bed volume. Fig. 6 shows the drainable porosities at a number of times. Due to the different column disassembly times, porosity values between each column

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**Fig. 5.** Variation in clog properties measured at column disassembly (a) total dry solids mass per bead versus column elevation, and (b) total volatile solids per bead versus column elevation. Note: Total dry solids = non-volatile solids (NVS) + volatile solids (VS).
section in Fig. 6 are the averaged values obtained from the columns remaining in operation when the test was performed. Where no value is shown, the drainable porosity was so low it could not be measured (less than 0.05).

During the first 13 days of column operation (6.7 m$^3$/m$^2$ of passed leachate), there was a uniform decrease in drainable porosity to 0.32 (16% decrease from the initial value), as shown in Fig. 6. It is likely that the FSS and microorganisms (VSS) within the leachate quickly adhered to the bead surface, thereby lowering the drainable porosity by (1) reducing the void volume, and (2) by changing the conditions for fluid retention and causing more leachate to adhere to the film/bead surface compared to the initially clean bead surface. However, the retention of suspended solids alone cannot explain the decrease in drainable porosity. Thus, part of the drainable porosity decrease must be due to biofilm growth resulting from the small removals of AA, PA, and BA (Fig. 3) during the first 13 days.

The 57-day (28 m$^3$/m$^2$ of passed leachate, near the end of the lag phase) average drainable porosity over the entire bed volume was 0.31 (19% decrease from the initial value), as shown in Fig. 6. However, the 57-day drainable porosity profile was no longer uniform with column elevation, and the bottom half of the column experienced a larger decrease in drainable porosity compared to the top half. Since there was greater removal of FSS and VSS at a similar time as the decrease in drainable porosity (most significantly within section S1), it appears that the partially clogged medium (i.e., larger particle diameter, smaller pore diameter) is more capable of retaining suspended solids than the unclogged medium. This is consistent with straining and filtration theories for removal of colloids in a porous medium (Tien, 1989; Indraratna, 1997; Reddi and Bonala, 1997). Generally, the straining and filtration of suspended solids is greater for smaller pore sizes (which correlates with lower drainable porosities) than for larger pore sizes (i.e., larger drainable porosities), assuming all other conditions are similar. Other factors such as the ionic strength of the leachate and interparticle forces (e.g., gravitational, inertial, hydrodynamic, electric double layer, and van der Waals) may also affect the removal of VSS and FSS from the leachate (Tien, 1989; Indraratna, 1997; Reddi and Bonala, 1997; Hajra et al., 2002). The lower drainable porosity in the bottom half of the column relative to the top half at 57 days is likely

### Table 1

<table>
<thead>
<tr>
<th>Units</th>
<th>Sample position (mm above column base)</th>
</tr>
</thead>
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<tr>
<td></td>
<td>0–100</td>
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<tr>
<td>Inorganic film density (Xf,i) (Mg NVS/m$^3$)</td>
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<tr>
<td>Volatile film density (Xf,a) (Mg VS/m$^3$)</td>
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<tr>
<td>Inorganic film thickness (Lf,i) (mm)</td>
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<tr>
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<tr>
<td>Effective porosity (calc) (–)</td>
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<tr>
<td>Drainable porosity (–)</td>
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<tr>
<td>Drainable porosity (–)</td>
<td>&lt;0.06</td>
</tr>
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</table>

(?) represents questionable measured results.
a result of (1) greater accumulation of suspended solids in the lower half of the column, and (2) larger amounts of biomass growth (BA degraders with some AA and PA degraders) in the lower half of the column, as indicated by the larger mass of dry and volatile solids within section S1 compared to sections S2–S6 at 57 days (Fig. 5).

4.2. Transition phase

During the transition phase of leachate treatment, there was a shift from a small removal of AA (55 days) to significant removal of AA (125 days), as shown in Fig. 4. The bulk of the AA removal occurred in the lower half of the column. BA removal at 125 days also occurred to a much greater extent in the lower half of the column than in the top half (Fig. 4) and more BA was removed at 125 days compared to 55 days at all column locations. Ca$^{2+}$ removal followed a similar trend as AA removal. PA was not significantly fermented during the transition phase.

The greater amounts of AA and BA removed at the end of the transition phase, compared to the end of the lag phase, indicates a substantial increase in microbial activity within the column during the transition phase. A larger amount of volatile solids was measured in the clog material at 115 days compared to 57 days at all column locations, with the largest increase in the lower half of the column (Fig. 5), confirming greater microbial activity at the latter time period. At 115 days there was about an order of magnitude larger mass of inorganic solids than volatile solids in the lower half of the column (Fig. 5). However, at this time, the inorganic film thickness was more than an order of magnitude smaller than the volatile film thickness (Table 1) due to the large contrast in inorganic and volatile film densities (discussed below). The increase in volatile and inorganic film thickness within the column caused the drainable porosity (over the entire bed volume) to decrease to a value of 0.26 (32% decrease from the initial value) at 115 days, with the largest decrease occurring within section S1 (43% decrease from the initial value), as shown in Fig. 6.

4.3. Steady state phase

During the steady state phase, >90% removal of AA and BA occurred within the column (Fig. 3). PA experienced some removal during the steady state phase. There was also about 40–60% removal of Ca$^{2+}$ during the steady state phase. At the end of the transition phase, there was a Ca$^{2+}$ removal of 60–70%. Since Ca$^{2+}$ removal is strongly influenced by AA fermentation (Rittmann et al., 1996), the lower percentage removal of Ca$^{2+}$ during the steady state phase compared to the transition phase is a result of lower influent AA concentration and
therefore lower mass of AA removed in the column during the steady state phase. During the 146-day (70 m$^3$/m$^2$ of passed leachate) drainable porosity measurement (and thereafter), the bottom 60 mm of the column could no longer be drained of fluid due to extensive clogging (Fig. 6). The average drainable porosity at the 146-day measurement time was less than 0.22 over the entire bed volume (42% decrease from the initial value and inferring a porosity of 0.20 within section S1). The final measured average drainable porosity measurement at 245 days (114 m$^3$/m$^2$ of passed leachate) was less than 0.15 over the entire bed volume, a 61% decrease in drainable porosity (inferring a porosity of 0.13 within sections S1 and S2).

The influent concentration of AA at 167 days was larger than at 245 days and the removal of AA at 167 days was greater than at 245 days, indicating that the percent removal of AA in the column is partly dependent on the influent concentration. This observation is consistent with theoretical considerations of substrate consumption within biofilms (Rittmann and McCarty, 2001). That is, the substrate flux within a biofilm is larger for higher concentrations of the substrate in the pore fluid, assuming all other factors are equal. Therefore, with respect to clogging of LCS, the larger the influent concentration of VFAs entering the collection system the greater the potential for substrate consumption. This results in greater calcium precipitation, biofilm development, and clogging.

The total dry and volatile solid masses within the pore space of the medium increased during the steady state phase (Fig. 5). Fig. 7 shows a photo of the clogged porous medium, at the influent end of the column, at disassembly after 245 days of operation. Note, the pore space between the porous medium visually appears to be filled with a solid black encrustation. The steady state phase inorganic film thickness was smaller than the volatile film thickness at all positions within the columns VK-3 and VK-4 (Table 1). Focusing on the influent end of the column (0–100 mm) at the end of the transition phase (VK-2 disassembly) until the final column was disassembled (VK-4) during the steady state phase (Table 1), the percentage of inorganic film thickness to total film thickness (sum of volatile and inorganic film thickness) increased from 21% at 115 days to 32% at 157 days, and then to 42% at 245 days. Since the inorganic (dry solids minus volatile solids) and volatile solid masses increased with time (Fig. 5), the percent increase in inorganic film thickness reflects a greater rate of mineral precipitation and FSS attachment compared to net biomass growth. Also, since AA, PA, and BA were fermented within the column, the total volatile solid (biomass) is likely partially comprised of AA, PA, and BA degraders, with each degrader likely having a different net growth rate (sum of biomass growth, biomass attachment from the passing leachate onto the medium surface, biomass loss due to detachment from the medium surface into the passing leachate, and biomass decay). Thus, the accumulation of volatile solid mass represents the net growth of each degrader, where each degrader may have a different net growth rate.

Biomass loss due to detachment has been subjected to some debate in literature. Rittmann (1982) suggested that biomass detachment from a medium surface is caused by fluid shear stress (i.e., mechanical stress) acting on the biofilm. Peyton and Characklis (1993) suggested that detachment may be a function of the growth rate of bacteria. If shear stress was the controlling mechanism for active biomass detachment, as the porosity of the medium decreases, there would be an increase in fluid seepage velocity and, therefore, larger fluid shear stresses acting on the biofilm causing greater detachment of biomass. Therefore, with longer elapsed times, the net biomass growth may be challenged by the greater detachment of biomass compared to shorter elapsed times due to the decrease in porosity with time.

Fig. 7. Photo of clog material and porous medium as observed at disassembly after 245 days of operation. Note: Pore space within the porous medium visually filled with a solid black encrustation.
Since the growth of each degrader (active biomass) and the accumulation of inorganic material cause a reduction in medium porosity (and larger shear stresses), both, the volatile and inorganic films, contribute to larger biomass detachment rates. This supports the increase in percent inorganic film thickness with time, as discussed above.

The volatile film density increased with time, with the largest increase near the influent end of the column (Table 1), where there was the largest decrease in porosity and therefore largest seepage velocity. Rittmann and McCarty (2001) stated that mechanical stresses on a biofilm tend to increase biofilm density and that high-stress anaerobic biofilms could reach densities to as much as 0.200 Mg VS/cm³. This is consistent with the active film density trends observed in this study.

5. Calcium in suspended solids

Field and laboratory studies (Brune et al., 1991; Fleming et al., 1999; Rowe et al., 2000a,b, 2002) showed that the clog solids contain high proportions of Ca²⁺ and CO₃⁻ constituents. The removal of dissolved Ca²⁺ from leachate has been shown (Rittmann et al., 1996) to be related to the fermentation of VFAs, primarily acetate (Fig. 4). Using the leachate chemistry results from this study, VanGulck et al. (2003) developed a method to predict biogeochemical precipitation of calcium carbonate due to the fermentation of VFAs. Colloidal chemistry studies on landfill leachate (Gounaris et al., 1993; Jensen and Christensen, 1998) showed that calcium could be present both in a dissolved and colloidal form. Thus, a fraction of the FSS may contain (1) calcium-bearing minerals suspended in leachate, and/or (2) calcium bound to suspended solids. Manning (2000) identified calcite, quartz, clay (kaolinite, illite, or chlorite), and bassanite and/or halite (CaSO₄·0.5H₂O) minerals in the suspended solids found in leachate. Additionally, calcium is probably a constituent of extracellular polysaccharides and/or proteins, which are used as linking materials in granulation of microorganisms (Morgan et al., 1991; Yu et al., 2001); thus, flocks of biomass may contain some calcium. Geochemical modeling studies have reported calcite and dolomite to be supersaturated in landfill leachate (Owen and Manning, 1997; Bennett et al., 2000; and Manning, 2000), indicating that precipitation of Ca²⁺ could occur, and also that the dissolution of bound calcium from the suspended solids is not a favorable reaction.

Assuming that particles smaller than 0.45 μm contribute to the dissolved phase particles (i.e., VSS), and particles larger than 0.45 μm contribute to the colloidal phase particles (i.e., FSS), the percentage of calcium in FSS (CaFSS) to FSS was deduced. The leachate studied by Gounaris et al. (1993) had a calculated percentage of CaFSS to FSS that ranged between 8% and 47%. The calculated percentage of CaFSS to FSS for four different leachates reported by Jensen and Christensen (1998) was 50%, 50%, 8%, and 20%. The percentage of CaFSS to FSS in KVL leachate ranged from 14% to 38%, with an average of 30% for six samples (McIsacc, personal communication). Knowing the cumulative mass of FSS retained within the column over the 245 day operating period, and using the 30% CaFSS to FSS measured in KVL leachate, the calculated CaFSS mass retained was about 11.6 g. From the total dry solids measured along the length of the column (Fig. 5) and the clog solid chemical composition (discussed below), the calcium mass precipitated after 245 days was calculated to be 43.2 g. Therefore, after 245 days of operation, there was about 3.7 times more calcium precipitated within the column than calcium retained by suspended solids. Since the concentration of FSS were deduced in the influent and effluent leachate only, the mass of this material retained and the distribution of CaFSS mass with column position is not known.

6. Hydraulic conductivity

The influent end of the column experienced the largest reduction in drainable porosity, and hence, the first measurable hydraulic conductivity decrease occurred after 108 days (55 m³/m² of passed leachate) for section S1 and 157 days (78 m³/m² of passed leachate) for section S2 within the column.

Rowe et al. (2000b) suggested an empirical relationship between the hydraulic conductivity, k, and drainable porosity, nₙ, for 6 mm glass beads:

\[ k = A n^{b n} \]

where A and b are coefficients from the regression analysis relating k and nₙ. As described earlier, the drainable porosity will yield values that are smaller than the actual porosity due to the inability to drain all the leachate out of the medium under a gravitational field at atmospheric pressure. The actual porosity, nₐ, can be estimated from the mass and bulk density measurements of the clog material at column disassembly (VanGulck, 2003), and this can then be correlated with the corresponding drainable porosity using data from numerous column experiments performed using 6-mm glass beads (Armstrong, 1998; Rowe et al., 2000a,b, 2002; and VanGulck, 2003). A regression line through the data points and forced through the initial (clean bead) actual and drainable porosity point (Fig. 8) gives:

\[ n_a = 0.75 n_d + 0.11. \]

This relationship has a correlation coefficient significantly different than zero at the 0.01 level. Applying this equation to the drainable porosities measured for this
experiment (shown in Fig. 6) and other column experiments (described above), a regression line was fitted through all the data points in the linear-log plot of actual porosity and hydraulic conductivity (Fig. 9) giving:

\[ k = 5.35 \times 10^{-10} e^{51.1 n_e} \] (m/s).

This relationship has a correlation coefficient significantly different than zero at the 0.01 level. The \( r^2 \) values for these relationships are low, due in part, to the fact that the type of clog material responsible for the decrease in \( n_d, n_e, \) and \( k \) changes with degree of clogging. As described above (and Table 1), at early stages of clogging...
the type of clog material is largely comprised of volatile mass, and at late stages of clogging the clog material is comprised on both volatile and inorganic mass. This relationship between hydraulic conductivity and actual porosity was developed from experiments completed with 6-mm diameter spheres and may not be universally applied to other porous media. There have been no other studies that have developed a relationship between these two parameters for various porous media types.

7. Clog solids composition

X-ray diffraction analysis completed on clog solids obtained from 25 to 75 mm above the base screen in columns VK-1 and VK-4 showed that magnesium-rich calcite (CaCO₃) was the dominant clog mineral (VanGulck, 2003). Magnesium-rich calcite has a calcite structure with some Mg²⁺ substituted for Ca²⁺ within the crystal lattice. A magnesium-rich calcite is different from dolomite (CaMg(CO₃)₂), which has a completely different crystal structure. The authors obtained similar results in the synthetic leachate column study (VanGulck, 2003). Thus, calcite precipitation within the column was the main contributor to the increase in inorganic film thickness deduced in this study. Calcite was also the dominant mineral in clog scale obtained from a leachate collection pipe in a Florida landfill that received incinerator ash and municipal solid waste (Maliva et al., 2000), and also in clog scale obtained from a United Kingdom leachate collection pipe (Manning, 2000). Thus, the mineral composition of the clog solids obtained in this study are consistent with that obtained in clog material from landfills.

Compositional analyses of clog solids were completed on samples retrieved from within (1) section S1 (clog sample location 0–25 mm above screen) for each column, (2) sections S5 and S6 (clog sample location 300–360 mm above the base screen) for VK-2, and (3) section S6 (clog sample location 300–325 mm above the base screen) for VK-4 (Table 2). The chemical composition of the clog solids (dry mass) during the lag phase had 9% calcium, 4% carbonate, 8% iron, and <1% magnesium. On a dry mass basis, the calcium and iron content within the clog material were similar at this time. During the steady state phase, the composition of the clog material averaged 28% calcium, 50% carbonate, 3% iron, and 1% magnesium on a dry mass basis. At the end of the transition phase and also during the steady state phase, the influent end of the column had marginally higher calcium (1–4%), lower carbonate (3%), and higher iron (2–3%) contents compared to the effluent end of the column. Comparing the clog solids composition within section S1 with time, the lag phase clog had much lower calcium and carbonate contents and a higher iron content, compared to the steady state clog.

The clog solids used for compositional analysis include Ca²⁺ and CO₃⁻ constituents from filtered/strained suspended solids (i.e., FSS), precipitated mass from the leachate (i.e., CaCO₃(s) precipitation due to VFA fermentation), and inert biomass. The contribution of Ca²⁺ and CO₃⁻ from inert biomass is likely small in comparison to that from FSS and precipitated mass. During the lag phase of COD removal, there was only a small amount of AA and Ca²⁺ removed within the column; therefore, the inorganic clog material during this time period is likely dominated by filtered/strained suspended solids, and yields a ratio of Ca²⁺/CO₃⁻ of 2.26. During the steady state phase, Ca²⁺ and CO₃⁻ constituents accumulated within the pore space of the column from filtered/strained suspended solids and precipitated mass, and had a ratio of Ca²⁺/CO₃⁻ of 0.56. If precipitation was the only mechanism responsible for Ca²⁺ accumulation within the clog material, and if all CO₃⁻ was bound to Ca²⁺, a mass balance consideration of CaCO₃(s) precipitation requires that the Ca²⁺/CO₃⁻ ratio be 0.666 (40 g Ca²⁺/60 g CO₃⁻). Completing a mass balance of calcium, and accounting for the Ca²⁺ bound to FSS, the calculated Ca²⁺/CO₃⁻ ratio for the precipitated material was 0.450 (VanGulck, 2003). Ratios less than 0.666 indicate other elements bound to CO₃⁻. Thus, 68% of the CO₃⁻ was bound to Ca²⁺ during the steady state phase. The remaining precipitated CO₃⁻ is likely bound to Fe²⁺, Mg²⁺, and Mn²⁺ (Table 2).

Comparing the clog composition measured in the field and column studies, the percentage of Ca²⁺ on a dry mass basis in the clog material differs; however, all studies show that Ca²⁺ and CO₃⁻ are the dominant constituents: 21%, 20%, 26%, 26%, 27%, and 28% Ca²⁺; 34, 30%, 50%, 52%, 49%, and 50% CO₃⁻; with 16%, 21%, 3%, 3%, 3%, and <1% Si reported by Brune et al. (1991), Fleming et al. (1999), Rowe et al. (2000a,b), Armstrong (1998), and this study (during steady state COD removal), respectively. The main difference between the field and laboratory clog composition is the Si content. The Si content in the field studies is likely the result of sand and silt particles from the waste and cover material being deposited in the drainage layer (Rowe et al., 2000a). In the laboratory, this material will have largely settled out before the leachate is passed through the column. If this material were present in the laboratory, it would comprise part of the FSS. However, the KVL column studies by Armstrong (1998) and Rowe et al. (2000a,b) found Si within the laboratory clog samples. The authors suggest that the Si content was due to the cutting procedure used to collect clog samples from within the column and not from the leachate itself. This current study did not employ the same cutting procedure to retrieve clog samples as in the Armstrong (1998) and Rowe et al. (2000a,b) studies, and as a result, there was negligible Si content within the clog. Rowe
et al. (2000a) showed that if an adjustment is made to exclude Si in the clog material, the percentage of Ca\(^{2+}\) in the field studies of 25%, based on Brune et al. (1991) and Fleming et al. (1999), is consistent with the column experiments permeated with KVL leachate of 26%, 26%, 27%, and 28%, based on Rowe et al. (2000a,b), Armstrong (1998), and this study (during steady state COD removal), respectively.

Observations by Fleming et al. (1999) during the KVL field exhumation, suggested that more clogging developed within areas of the collection system where there was no geotextile placed between the stone and waste, compared to areas with a geotextile. The KVL clog composition results reported in Fleming et al. (1999) were obtained from an area of the collection system with no filter/separator between the waste and drainage stone. If an appropriate filter/separator was used to separate the waste and stone, the Si content in the leachate as it enters the drainage stone would likely be small, and hence, the percentage of Si in the clog material would be small. Thus, the time to clog a drainage layer without a filter/separator would likely be quicker than one with a filter/separator, since less material (i.e., sand particles) could enter and accumulate within the drainage layer, assuming all other factors are similar.

Rowe and Fleming (1998) (see also Rowe, 2001) developed a technique to predict the time to clog LCS. One of the key input variables is the fraction of Ca\(^{2+}\) in clog material (\(f_{\text{Ca}}\)). Comparing the \(f_{\text{Ca}}\) obtained in field (20%) to the column experiments permeated with KVL leachate (25–31%) provides some guidance on the selection of this value in predicting the service life of collection systems with and without a filter/separator, respectively.

### 8. Practical implications

The results of this study showed that a gravel-size material permeated with leachate, can become clogged over time due to the accumulation of volatile and inorganic materials within the pore spaces of the porous medium. The development of a biofilm occurs relatively quickly, and that, with time, this biofilm changes from a soft ‘slime’ to a slime with hard particles (sand-size solid material in a soft matrix), to a solid porous concretion of a coral-like ‘biorock’ structure. During the early phases of development of the active biofilm, it would be relatively easy to clean from leachate collection pipes (e.g., by pressure jetting). However, as the biofilm passes from the transition to the steady state phase, the inor-

### Table 2

Compositions of clog material within KVL leachate column experiments (nd- below detection limit)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Units</th>
<th>VK-1 S1 57 days</th>
<th>VK-2 S1 115 days</th>
<th>VK-3 S1 157 days</th>
<th>VK-4 S1 245 days</th>
<th>VK-2 S5&amp;S6 115 days</th>
<th>VK-4 S6 245 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water content</td>
<td>%wet</td>
<td></td>
<td>76.63</td>
<td>54.6</td>
<td>44.7</td>
<td>30.5</td>
<td>76.7</td>
<td>48.5</td>
</tr>
<tr>
<td>Organic carbon</td>
<td>C</td>
<td>%</td>
<td>16.7</td>
<td>–</td>
<td>5.7</td>
<td>4.6</td>
<td>–</td>
<td>1.6</td>
</tr>
<tr>
<td>Organic matter (TVS)</td>
<td>%</td>
<td></td>
<td>29.7</td>
<td>15.3</td>
<td>10.5</td>
<td>8.6</td>
<td>20.4</td>
<td>11.4</td>
</tr>
<tr>
<td>Calcium</td>
<td>Ca</td>
<td>%</td>
<td>8.6</td>
<td>21.0</td>
<td>27.9</td>
<td>28.7</td>
<td>17.2</td>
<td>28.4</td>
</tr>
<tr>
<td>Carbonate as CO(_3)</td>
<td>CO(_3)</td>
<td>%</td>
<td>3.8</td>
<td>–</td>
<td>44.2</td>
<td>48.7</td>
<td>–</td>
<td>52.0</td>
</tr>
<tr>
<td>Ca/CO(_3)</td>
<td></td>
<td></td>
<td>2.26</td>
<td>0.63</td>
<td>0.59</td>
<td>–</td>
<td>0.55</td>
<td></td>
</tr>
<tr>
<td>Aluminum</td>
<td>Al</td>
<td>%</td>
<td>0.196</td>
<td>0.056</td>
<td>0.037</td>
<td>0.016</td>
<td>0.257</td>
<td>0.111</td>
</tr>
<tr>
<td>Carbon</td>
<td>C</td>
<td>%</td>
<td>18.6</td>
<td>17.2</td>
<td>14.8</td>
<td>14.1</td>
<td>23.7</td>
<td>15.3</td>
</tr>
<tr>
<td>Iron</td>
<td>Fe</td>
<td>%</td>
<td>7.851</td>
<td>4.847</td>
<td>3.588</td>
<td>3.144</td>
<td>2.179</td>
<td>2.630</td>
</tr>
<tr>
<td>Magnesium</td>
<td>Mg</td>
<td>%</td>
<td>0.384</td>
<td>0.546</td>
<td>0.882</td>
<td>1.098</td>
<td>0.384</td>
<td>1.356</td>
</tr>
<tr>
<td>Manganese</td>
<td>Mn</td>
<td>%</td>
<td>0.152</td>
<td>0.250</td>
<td>0.266</td>
<td>0.245</td>
<td>0.219</td>
<td>0.165</td>
</tr>
<tr>
<td>Potassium</td>
<td>K</td>
<td>%</td>
<td>0.353</td>
<td>0.172</td>
<td>0.105</td>
<td>0.038</td>
<td>0.105</td>
<td>0.075</td>
</tr>
<tr>
<td>Silicon</td>
<td>Si</td>
<td>%</td>
<td>0.733</td>
<td>0.252</td>
<td>0.201</td>
<td>0.126</td>
<td>0.560</td>
<td>0.215</td>
</tr>
<tr>
<td>Sodium</td>
<td>Na</td>
<td>%</td>
<td>0.623</td>
<td>0.307</td>
<td>0.193</td>
<td>0.189</td>
<td>0.282</td>
<td>0.233</td>
</tr>
<tr>
<td>Sulfur</td>
<td>S</td>
<td>%</td>
<td>7.25</td>
<td>2.8</td>
<td>1.04</td>
<td>0.7</td>
<td>0.55</td>
<td>0.37</td>
</tr>
<tr>
<td>Titanium</td>
<td>Ti</td>
<td>%</td>
<td>0.026</td>
<td>0.008</td>
<td>0.001</td>
<td>nd</td>
<td>0.077</td>
<td>0.011</td>
</tr>
<tr>
<td>Total phosphorus</td>
<td>P</td>
<td>%</td>
<td>0.078</td>
<td>0.083</td>
<td>0.055</td>
<td>0.044</td>
<td>0.044</td>
<td>0.037</td>
</tr>
<tr>
<td>Barium</td>
<td>Ba</td>
<td>ppm</td>
<td>53</td>
<td>81</td>
<td>96</td>
<td>103</td>
<td>63</td>
<td>110</td>
</tr>
<tr>
<td>Boron</td>
<td>B</td>
<td>ppm</td>
<td>40.64</td>
<td>–</td>
<td>37.95</td>
<td>10.64</td>
<td>36.89</td>
<td></td>
</tr>
<tr>
<td>Cadmium</td>
<td>Cd</td>
<td>ppm</td>
<td>0.252</td>
<td>–</td>
<td>0.129</td>
<td>0.341</td>
<td>0.129</td>
<td></td>
</tr>
<tr>
<td>Chromium</td>
<td>Cr</td>
<td>ppm</td>
<td>14</td>
<td>–</td>
<td>15</td>
<td>19</td>
<td>14</td>
<td></td>
</tr>
<tr>
<td>Cobalt</td>
<td>Co</td>
<td>ppm</td>
<td>29.5</td>
<td>–</td>
<td>10</td>
<td>12.4</td>
<td>6.8</td>
<td></td>
</tr>
<tr>
<td>Copper</td>
<td>Cu</td>
<td>ppm</td>
<td>196.3</td>
<td>–</td>
<td>72.90</td>
<td>432.4</td>
<td>269.1</td>
<td></td>
</tr>
<tr>
<td>Lead</td>
<td>Pb</td>
<td>ppm</td>
<td>21.32</td>
<td>–</td>
<td>8.027</td>
<td>28.71</td>
<td>15.17</td>
<td></td>
</tr>
<tr>
<td>Molybdenum</td>
<td>Mo</td>
<td>ppm</td>
<td>0.152</td>
<td>0.250</td>
<td>0.266</td>
<td>0.245</td>
<td>0.219</td>
<td>0.165</td>
</tr>
<tr>
<td>Nickel</td>
<td>Ni</td>
<td>ppm</td>
<td>211.4</td>
<td>–</td>
<td>69.53</td>
<td>91.0</td>
<td>50.24</td>
<td></td>
</tr>
<tr>
<td>Strontium</td>
<td>Sr</td>
<td>ppm</td>
<td>183</td>
<td>492</td>
<td>707</td>
<td>773</td>
<td>371</td>
<td>856</td>
</tr>
<tr>
<td>Zinc</td>
<td>Zn</td>
<td>ppm</td>
<td>593.2</td>
<td>200.3</td>
<td>579.1</td>
<td>337.4</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
ganic film becomes firmly attached to the adjacent media (beads in this case, but a similar behavior can be expected on the perforation in pipes and on the inside of pipes (see Fleming et al., 1999). At this stage, it would be very difficult to remove the clog material. There are five important practical implications to this.

Firstly, leachate collection pipes in landfills should be cleaned of bioslime on a regular basis to remove the soft biofilm in its early stages of development. Secondly, it has been observed that the rate of clog development accelerates with time and hence the rate of cleaning required in pipes may increase as the concentration of VFAs in the leachate (or COD) increases. Thirdly, since the carbonate in the clog arise from biological processes (Rittmann et al., 1996; Bennett et al., 2000; VanGulck et al., 2003), the control on clogging is due to the supply of silica, calcium, and VFAs. Silica (sand and silt particles) intrusion can be controlled by the use of an appropriate filter (Rowe, 2001), however, the filter itself may clog due to biological, chemical, and physical processes. Calcium and VFAs are ubiquitous in waste and cannot be controlled as such. However, it should be recognized, the management practice will impact the VFAs reaching the leachate collection system. Armstrong and Rowe (1999) discuss issues related to waste placement. Also, it should be recognized that practices such as recirculation of leachate serve to re-inject VFAs and calcium back into the waste, and this will have implications for the performance of the leachate collection system. It is also important to note that VFA concentrations are highest in young refuse, and that as the refuse decomposes, VFA concentrations may decrease. In this study, a “young” leachate was used. The rate of clog development may be different if an “old” leachate, with low VFA and Ca$^{2+}$ concentrations, had been used. Fourthly, it is noted that, due to the level of “treatment” that occurs in a leachate collection system before the leachate reaches the collection point, analysis of the leachate VFA and Ca$^{2+}$ concentrations only indicate what it is like at the collection point and provide little evidence regarding the nature of the leachate that enters the system. Finally, since clog material accumulates heavy metals as well as Ca$^{2+}$ and Mg$^{2+}$, techniques such as acid flushing to treat developed clog material have the potential to generate a leachate with higher concentration of metals. Thus, it is likely to be preferable to flush with water while the film is soft rather than trying to clean up the clog after it has entered the solid phase of development.

9. Conclusions

Through controlled column experiments containing a gravel-size medium permeated with KVL leachate, a reduction in medium porosity (and hydraulic conductivity) was caused by (1) the fermentation of VFAs, (2) precipitation of cations, primarily Ca$^{2+}$, out of solution, and (3) retention of suspended solids. Thus, three distinct clogging mechanisms (biological, chemical, and physical processes) resulted in a $\geq 60\%$ reduction in drainable porosity and a six-order magnitude decrease in hydraulic conductivity. These three clogging mechanisms contributed to the accumulation of volatile (biomass) and an inorganic (mineral precipitate and retained non-active suspended solids) material within the pore space of the medium. The data suggests that the contribution of one of the clogging processes influences the other two, and therefore, the rate of clogging. For example, retention of volatile solids increases the biomass within the column, which could then increase the amount of VFAs consumed, and therefore increases the amount of Ca$^{2+}$ precipitated. This increase in biomass and precipitated Ca$^{2+}$ reduces the medium porosity, and therefore affects the retention of suspended solids and decreases the rate of VFA fermentation due to a lower retention time of leachate within the column.

Microorganisms suspended in the KVL leachate colonized the gravel-size material within the column. These microorganisms grew as a surface biofilm capable of removing organic acids in the passing leachate, leading to biological clogging. The development of the biofilm was influenced by (1) growth rate of the biomass attached to the medium and (2) retention of biomass (VSS) from the passing leachate onto the medium. Comparing two similar columns, one permeated with synthetic leachate that contained negligible suspended solids, and one with real leachate (this study), the retention of VSS was largely responsible for the development of a sufficiently thick biofilm capable of removing about 10% of the influent COD. Of the three primary VFAs in KVL leachate, butyrate degraded before acetate, and acetate degraded before propionate.

The changes in leachate composition with time and position in the column highly suggests that fermentation of acetate was the driving force for the precipitation of dissolved calcium from the leachate. The clog solids were primarily composed of Ca$^{2+}$ and CO$_3^{2-}$, and 68% of the CO$_3^{2-}$ precipitated from the leachate is bound to Ca$^{2+}$ as CaCO$_3(s)$ during steady state leachate treatment. The remaining CO$_3^{2-}$ is likely bound to Fe$^{2+}$, Mg$^{2+}$, and Mn$^{2+}$.

In addition to the precipitation of minerals, the retention of FSS from the leachate onto the medium surface contributed to the inorganic clog mass. Since FSS are partially comprised of Ca$^{2+}$, the clog solids Ca$^{2+}$ content contains (1) Ca$^{2+}$ deposited as a result of straining/filtration of FSS and (2) dissolved Ca$^{2+}$ precipitating as CaCO$_3(s)$ due to VFA fermentation. Over the duration of the experiment, 3.7 times more Ca$^{2+}$ was precipitated in the column than Ca$^{2+}$ was retained with FSS retention.
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Testing and Materials, West Conshohocken, PA, USA.

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