

PARTICLE SIZE AND CLOGGING OF GRANULAR MEDIA PERMEATED WITH LEACHATE

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ABSTRACT: The effect of particle size (4-, 6-, and 15-mm nominal sizes) on the rate of clogging of columns of porous media permeated with municipal solid-waste leachate is examined. Clogging is shown to be more localized over a small volume of the porous media near the influent end of the column for smaller particles than for larger particles, where clogging was more uniformly distributed along the column. This is attributed to the greater surface area per unit volume of smaller particles allowing greater biofilm growth per unit volume. This increased the reduction in chemical oxygen demand (COD) and caused greater deposition of inorganic clog material per unit length of column than for larger particles. The distribution of methanogenic bacteria was found to closely correspond to the zones of most severe clogging. The bulk density of clog material is shown to be between 1.6 and 1.8 Mg/m³. The chemical composition of the clog material is essentially independent of particle size, with calcium representing 26% of the dry mass of the clog material and CaCO₃ being the main component of the clog. An examination of the yield of CaCO₃ relative to COD indicates that the carbon in the CaCO₃ represents <4% of the organic carbon represented by the drop in COD. Finally, the data from the column test is used to predict the expected time to clog for an actual landfill and were found to give results consistent with what was observed in the field.

INTRODUCTION

Most modern municipal solid waste (MSW) landfills have a leachate collection system (LCS) as part of the engineered barrier design. The characteristics of MSW leachate are such that there is a potential for biologically induced clogging of the LCS (Rowe et al. 1995). The work by Brune et al. (1994) and Rittmann et al. (1996) outlined processes for the accumulation of clog material in the drainage layer. There is some evidence that the particle size of the drainage media may influence the time required for an LCS to fail due to clogging. The importance of this design factor with respect to North American landfills has been highlighted by observations at two landfills discussed in the following paragraphs.

The first landfill has an LCS constructed of a continuous 0.3-m drainage blanket of relatively uniformly graded (50-mm nominal size) crushed stone with leachate collection pipes spaced about 200 m apart. Field exhumations were carried out for sections of the LCS that had been exposed to leachate for periods of 1–4 years (Fleming et al. 1999). The accumulation of clog material (that included both organic and inorganic material) was greatest near the leachate collection pipe. Although there was a significant reduction in void space due to the accumulation of clog material, there was no significant leachate mound and the LCS was still performing adequately at these locations. They found that approximately 50% of the clog material at this landfill was calcium carbonate (CaCO₃), thus identifying a need to focus on CaCO₃ deposition in clogging studies.

The second landfill has an LCS consisting of a 200-mm-diameter, perforated, corrugated steel (16-gauge) pipe, surrounded by 9-mm-diameter pea gravel (extending to a radius of about 0.5 m from the bottom of the pipe), at a spacing of 50 m (newer portions) to 200 m (older portions) on a relatively

flat base. Because the grading of the pea gravel is unknown, the D_{10} is unknown; however, it is expected to lie between 4 and 15 mm (the range of particle size examined in this paper). A significant leachate mound had developed in the area of the landfill with the LCS described above (Rowe 1998). This mound first became evident about 11 years after landfilling commenced. After 16 years the leachate mound was about 23 m above the base of the landfill and the LCS was only collecting about 6% of the estimated fluid input. Because the waste deposited at the two landfills was similar, one would expect the initial leachate characteristics to be similar. The only notable difference between the waste at the two landfills is the presence of sewage sludge, representing about 10% of the waste deposited at the second landfill.

The difference in behavior of these two landfills raises some questions with respect to the effect of the differences in the size of drainage media used in the LCS. The smaller particle diameter of the granular media around the collection pipes at the second site may have substantially decreased the pore size around the collection pipes, in contrast to the first site. Two granular materials with similar packing factors may have similar porosities, yet the sample with the larger D_{10} will have the larger pore size. One can hypothesize that a decrease in particle diameter increases the rate of clogging around leachate collection pipes; however, it would be highly desirable to have experimental evidence to substantiate the effect of particle diameter on the rate of clogging of porous media.

Prior to serious clogging of typical underdrain systems that incorporate granular blanket drains, the flow is largely vertical through the unsaturated zone in the upper portion of the blanket drain and predominantly horizontal in the lower saturated zone, where clogging has been observed to be greatest [e.g., Fleming et al. (1999)]. Thus, with respect to predicting clogging, the zone of greatest interest and concern is the saturated zone, where there is typically the greatest flow and mass loading, and this will be the focus of this paper.

The primary objective of the experimental study reported herein is to examine the effect of the particle diameter on the rate of clogging in controlled laboratory column experiments using leachate collected from the first landfill site. The columns were designed using granular materials with a D_{10} size that bracketed the expected diameter for the pea gravel used around the pipe at the second landfill and a flow that was expected at the second landfill prior to clogging. A second objective is to provide experimental parameters, obtained un-

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der controlled conditions, that can be used in the approximate engineering approach for estimating the rate of clogging of LCS proposed by Rowe and Fleming (1998). A third and final objective is to compare the predicted time of clogging of the collection system for the second landfill (based on experimental data in the laboratory) with that observed in the field. This study provides the first published data relating reduction in drainable porosity (due to biologically induced chemical clogging) to the change in hydraulic conductivity for different sizes of particles.

A separate series of mesocosm tests, which simulate at full scale and in real time both the top-down unsaturated flow and the horizontal saturated flow as one approaches the leachate collection pipe (Rowe et al. 1997), have been running continually since June 1993 and will be reported after termination.

RELATED STUDIES

Brune et al. (1994) described results from a laboratory research program involving columns packed with granular material with particle sizes in the ranges (smallest–largest) 2–4, 4–8, 8–16, and 16–32 mm. At the conclusion of testing (between 406 and 462 days), it was found that the absolute reduction in pore space was about 20% for the 2–4 and 4–8 mm grain-size columns, which was two to three times greater than the 8–16 mm grain-size columns and four times greater than the 16–32 mm grain-size columns. These results suggest that the finer-grained material is more susceptible to clogging than coarse-grained materials. Unfortunately, no attempt appears to have been made to relate the clogging to leachate chemistry, or to evaluate the change in hydraulic conductivity with time or the time to clog.

Peeling et al. (1999) and Paksy et al. (1998) conducted tests using columns packed with Thames gravel with particle diameter ranges of 5–10, 10–20, and 20–40 mm with D_{10} of 5.8, 10, and 22 mm, respectively. Synthetic leachate was pumped through at rates comparable to annual percolation rates ($0.2 \text{ m}^3/\text{m}^2/\text{a}$, where a = annum, or 0.57 L/day). After column operations of 700 days, it was found that the reduction in drainable porosity averaged over the whole bed volume had been reduced by 7, 4, and 1% for the effective particle sizes 5.8, 10, and 22 mm, respectively. The small decrease in drainable porosity was likely due to the low concentrations of calcium and iron (200 and 100 mg/L, respectively) in their synthetic leachate, which were much lower than the concentrations in the real Keele Valley Landfill leachate used in the present tests.

METHODOLOGY

The testing involved passing leachate upward through columns containing glass beads with nominal diameters of 4, 6, and 15 mm. Glass beads were used in these experiments for two reasons. First, so that the effect of particle size would be clear, a truly uniform media was required. By using glass beads it was possible to ensure that all “4-mm beads” were in fact 4 mm in diameter and all “6-mm beads” were 6 mm in diameter, whereas for real granular material it is difficult to obtain this level of consistency. Second, it was considered important to have tests with well-defined particle sizes in order to be of most use in validating clogging models that typically assume a regular particle size [e.g. Rowe et al. (1997) or Cooke et al. (1999)]. Tests are presently being performed using pea gravel to allow a comparison of the behavior of glass beads with that of gravel having a similar nominal size. When completed, the results from these tests will be reported. All columns were operated at a temperature of $27^\circ \pm 1^\circ\text{C}$ that approximates the temperatures of the leachate at the second landfill discussed earlier [$27^\circ\text{--}32^\circ\text{C}$, Rowe (1998)]. Six tests (denoted PK1, PK2, . . . , PK6) were performed under saturated conditions. Columns PK1 and PK2 had 4-mm beads and were terminated after 146 and 252 days, respectively. Columns PK3 and PK4, with 6-mm beads, were terminated after 257 and 265 days, respectively. Finally, columns PK5 and PK6, with 15-mm beads, were terminated after 383 and 422 days, respectively.

Each column was about 0.7-m long, with an inlet at the bottom and outlet at the top. Flow was from bottom to top to more readily maintain saturated conditions similar to those experiences at the bottom of LCSs. Five piezometers were located along the column, as shown in Fig. 1. Anaerobic conditions within the columns were maintained at all times, and excess gas was collected in Tedlar gas collection bags (Fig. 1). The piezometers also were connected to the gas system (Fig. 1). The flow rate was specified (and monitored).

The initial hydraulic conductivity was measured using a constant head test, but this approach could not be used once permeation with leachate had begun. During leachate permeation, it was possible to infer the change in hydraulic conductivity with time in the four regions between piezometers by measuring the head in piezometers P1–P5. However, because the flow rate ($\cong 6 \times 10^{-6} \text{ m/s}$) was small during permeation, the hydraulic conductivity between piezometers could not be accurately measured until the material had clogged sufficiently

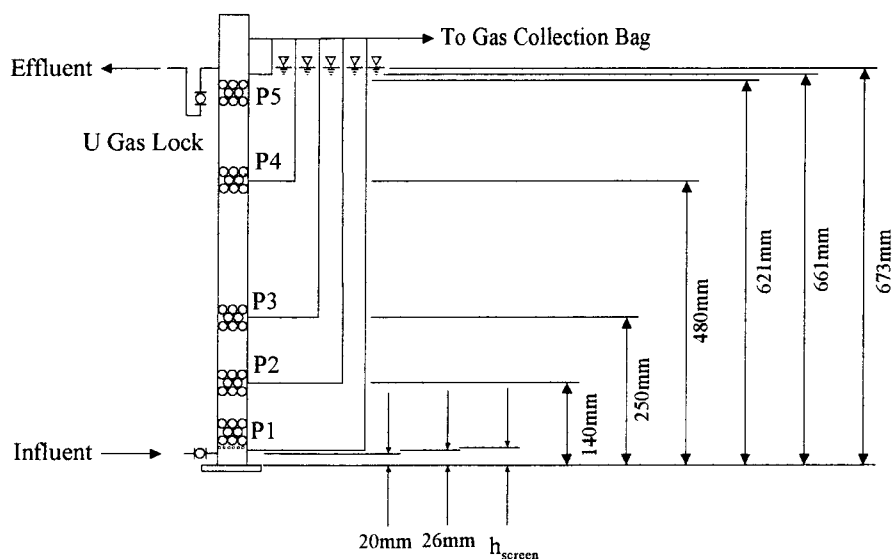


FIG. 1. Column Design Showing Typical Dimensions

to cause a measurable difference in head between two piezometers. Due to the need to maintain anaerobic conditions and not disturb the biofilm and the effect of small gas bubbles that formed in the piezometer tubing, it was not practical to detect changes in hydraulic conductivity until it had dropped by about 5 orders of magnitude.

Periodically, the flow was stopped and the drainable porosity was measured by allowing leachate to slowly drain out of the column. Anaerobic conditions were maintained during drainage by keeping the system connected to a Tedlar gas collection bag containing landfill gas generated by the column. The amount of water drained out between two specified locations in the column was measured and the “drainable porosity” between these two elevations was deduced. This drainable porosity represents the pore space that will freely drain in a gravitational field at atmospheric pressure and hence represents the porosity readily available for leachate flow. It is acknowledged that this does not include pore space occupied by fluid retained on the solid particles and held in small pores within the clog structure by capillarity or blockage. Thus, the drainable porosity is less than the actual porosity but is representative of the porosity of primary relevance to leachate flow in an LCS. It was generally not practical to measure drainable porosities of <5%.

The MSW leachate used in the test was regularly collected from the Keele Valley Landfill, in Maple, Ontario, Canada. This leachate has already passed through an LCS and may not be typical of actual leachate entering an LCS, because there will have been consumption of volatile fatty acids [and hence a decrease in chemical oxygen demand (COD)] and precipitation of CaCO_3 as it moved through the granular material before reaching the collection pipes. However, this leachate was considered to be reasonably representative of the leachate in the granular material adjacent to the leachate collection pipes, where the greatest clogging was observed to occur by Fleming et al. (1999). Thus the columns are most representative of the saturated zone within 0.5 m of the leachate collection pipe. The leachate was transported to the laboratory in sealed high-density polyethylene tanks (to maintain anaerobic conditions) and stored at 10°C until used. The average concentrations of COD and CaCO_3 were 11,000 mg O_2/L (range 8,410–13,970 mg O_2/L) and 1,360 mg/L (range 910–1,890 mg/L), respectively. The total suspended solids in the influent leachate ranged between 161 and 5,025 mg/L, with an average of 860 mg/L. The leachate was passed through the column once and was not recycled.

Leachate was fed into each column pair with a peristaltic pump at a rate to give an average Darcy flux of $0.51 \text{ m}^3/\text{m}^2/\text{day}$ ($\cong 6 \times 10^{-6} \text{ m/s}$ or 1 l/day for each column), which was selected to approximate the expected flow rate in the saturated pea gravel adjacent to the collection pipes at the second landfill (prior to excessive clogging). This is based on an average drainage path $L = 100 \text{ m}$, a typical leachate generation rate for the region of $q_0 = 0.1 \text{ m}^3/\text{a}/\text{m}^2$ of the plan area, and a saturated thickness of $h = 0.05 \text{ m}$. The flow rate in each column was monitored and every effort was made to maintain a flux of approximately $0.51 \text{ m}^3/\text{m}^2/\text{day}$ (1 l/day for these columns); however, as shown in Fig. 2, for each pair of columns there was some variability in flow rate. Although this was a result of experimental challenges, the fluctuations shown in Fig. 2 are also typical of what is observed in the field (Armstrong and Rowe 1999), and it is the average and accumulated flow that can be expected to control the clogging of the system. Eventually, the columns became sufficiently clogged so that the flow rate could no longer be maintained, and at this point the tests were terminated.

The influent and effluent were tested for COD, hardness as CaCO_3 , pH, E_h , and fluid temperature on a weekly basis. To

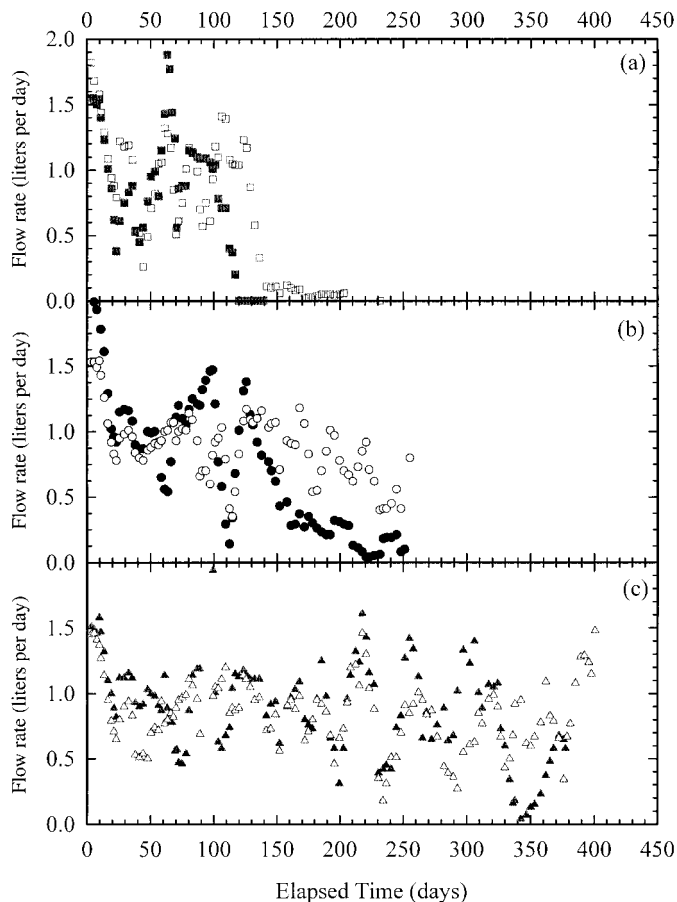


FIG. 2. Flow Rate with Respect to Time: (a) 4-mm Bead Columns; (b) 6-mm Bead Columns; (c) 15-mm Bead Columns

ensure that the columns were operating at their design flow rates, the flow rates were checked twice per week. The drainable porosity and hydraulic conductivity were monitored under anaerobic conditions. Biological activity was monitored by performing Biological Activity Reaction Tests (BART) (Cullimore 1999).

At the conclusion of the testing for each column, the columns were placed in a glove box with a nitrogen atmosphere (to maintain anoxic conditions) and the glove box was cut open to allow access to the beads. Beads were taken from six sampling points (25–100, 100–200, 200–300, 300–400, 400–500, and 500–600 mm above the column base). The biofilm properties were measured by weighing the samples and drying them at 105°C to establish the baseline biofilm water content per bead. The beads were then weighed again, heated to 550°C and reweighed to obtain the mass of organic material per bead. Finally, the glass beads were cleaned to obtain the mass of inorganic material per bead. The biofilm was measured for the wet and dry densities. Samples of dry clog material was sent for elemental analysis.

CHANGES IN LEACHATE AS IT PASSED THROUGH COLUMNS

Fig. 3 shows the variation in normalized (effluent concentration divided by influent concentration) COD and CaCO_3 concentrations with time in all six tests. There was about a 20-day time lag between test initiation and CaCO_3 removal for all columns. This lag is considered to be due to time required to establish a biofilm on the beads that will “treat” the leachate. During the first 20 days there was only a minor reduction in normalized COD, a modest shift in pH (from about pH 6.6 to 6.9), and no significant change in the normalized CaCO_3

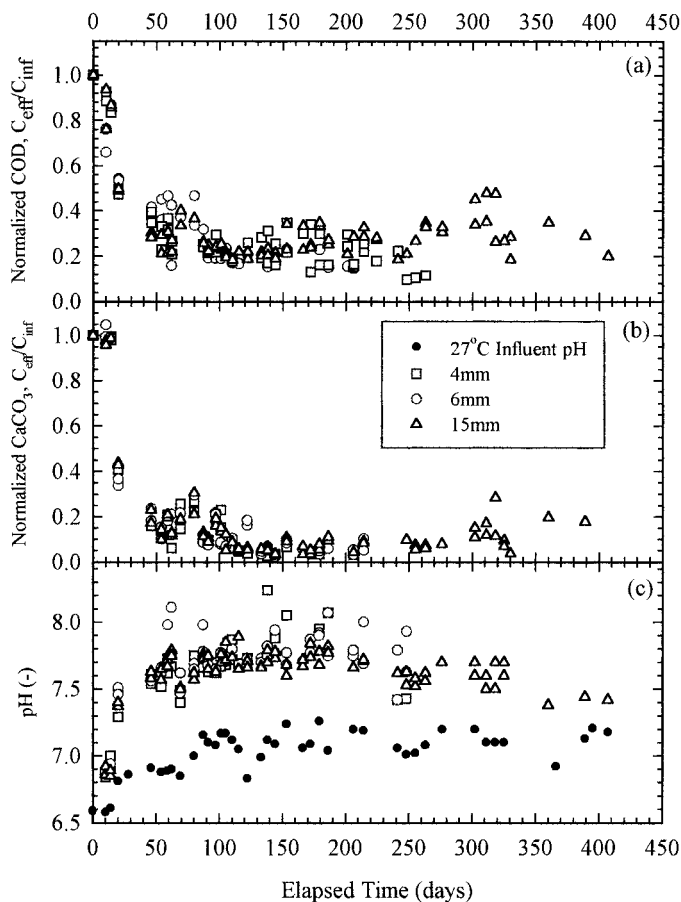


FIG. 3. Effect of Particle Size on: (a) Normalized COD with Elapsed Time; (b) Normalized CaCO₃ with Elapsed Time; (c) pH versus Elapsed Time

concentration that remained at $[\pm 0.05]$, Fig. 3]. Between 20 and 140 days, the normalized COD dropped to about 0.25, the pH of the effluent moved to about pH 7.8–7.9, and the normalized CaCO₃ concentration dropped to <0.05 . This suggests that the general trend in terms of COD consumption and deposition of CaCO₃ were practically independent of the particle diameter over the range examined. However, as will be demonstrated in subsequent sections, the columns packed with 4-mm-diameter particles experienced a significant reduction in drainable porosity and hydraulic conductivity much earlier than the 6-mm-bead columns, which in turn clogged faster than the 15-mm-bead columns. For each test, the hydraulic conductivity eventually dropped sufficiently enough that it was no longer practical to maintain the design flow rate and the test was terminated. When the tests were terminated, the volume of leachate through the columns averaged 68, 99, and 161 m³/m² for the particle diameters of 4, 6, and 15 mm, respectively.

Comparing the different volumes of leachate required to cause clogging, it can be appreciated that, on average, the 6-mm beads took 50% longer to clog than the 4-mm beads, and the 15-mm beads were about two to three times longer to clog than the 4-mm beads and 60% longer to clog than the 6-mm beads. As discussed in a later section, these numbers can be used to estimate the time required to clog the drainage material around a leachate collection pipe and, as will be seen, correspond to between 5 and 13 years of leachate flow into a pipe at landfill 2.

CALCIUM CARBONATE YIELD

The yield Y_C of CaCO₃ was obtained by plotting the COD removed versus the CaCO₃ removed and fitting the data by

linear regression. Due to scatter in the data (Fig. 4), the r^2 value was low; however, the data does indicate a trend with CaCO₃ removed increasing with increasing COD removed. The yield coefficients were calculated for each particle size and found to be 0.15, 0.14, and 0.15 mg CaCO₃/mg COD for the 4-, 6-, and 15-mm-bead columns, respectively. These are very similar, and an analysis of variance (ANOVA) test for the three data sets indicated that the differences are not statistically significant. When all the data are combined, the yield coefficient is approximately 0.15 CaCO₃/mg COD.

The yield coefficient can be considered in terms of carbon held in the calcium carbonate versus carbon from the COD. Consider that carbon accounts for 0.12 mg/mg CaCO₃, and the yield coefficient is approximately 0.15 mg CaCO₃/mg COD, which gives 0.018 mg C in CaCO₃/mg COD. The COD of the Keele Valley leachate is primarily comprised of acetic, propionic, and butyric acids and, assuming that the recalcitrant portion of the COD is evenly distributed between the three primary components, there is approximately 0.46 mg C/mg COD. Thus the carbon in the carbonate deposited within the columns would account for approximately 4% of the total carbon removed from the leachate, in terms of COD removed. The remainder of the carbon is primarily released as by-products of microbiological processes in the form of carbon dioxide and methane, which was collected in Tedlar gas bags but not monitored for mass balance.

BART RESULTS

The BART were performed on fluid samples during normal column operation and on solid clog material as part of an autopsy procedure after termination of a column. The BART can be used to indicate the microbial community structure from reaction patterns. The probable population can be inferred from the measured time delay between test initiation and first reaction. These tests were used to define the level of aggressivity that is a measure of relative population and microbial community structure within the columns. The biodefectors used during the course of this study were capable of identifying the presence/absence and relative aggressivity of heterotrophic aerobic bacteria, particularly those that are facultative anaerobes; iron oxidizing and reducing bacteria (IRB), including sheathed iron bacteria such as *Gallionella*, pseudomonads, and enterics; sulfate-reducing bacteria (SRB); slime-forming bacteria (SLYM); denitrifying bacteria capable of reducing nitrate to nitrogen gas (DNs); total coliforms; and methanogenic bacteria (BIOGAS). Full details are given by Armstrong (1998). The following provides a summary of the findings from the microbiological tests.

Both the influent and effluent samples consistently contained a very aggressive microbial community (large population) dominated by facultative anaerobes. Over the course of the testing, there was a decrease in the population of iron-related bacteria in the leachate influent while the effluent population remained relatively constant. This suggests that iron-related bacteria are growing within the columns and the effluent samples contain bacteria that have detached from the biofilm. At column initiation a diverse sulfate-reducing community was present in the leachate and, over the course of the testing, there was also a reduction in the influent aggressivity (population) for SRB. Initially, the effluent was similar to the influent in terms of both community and aggressivity but, with time, the effluent aggressivity (population) increased. This is attributed to growth and detachment of SRB that increased until the end of the tests. The leachate testing indicates that a mixed community of facultative anaerobes, iron-related bacteria, SRB, slime formers, and enterics were present within all of the columns. From the leachate data there was no apparent

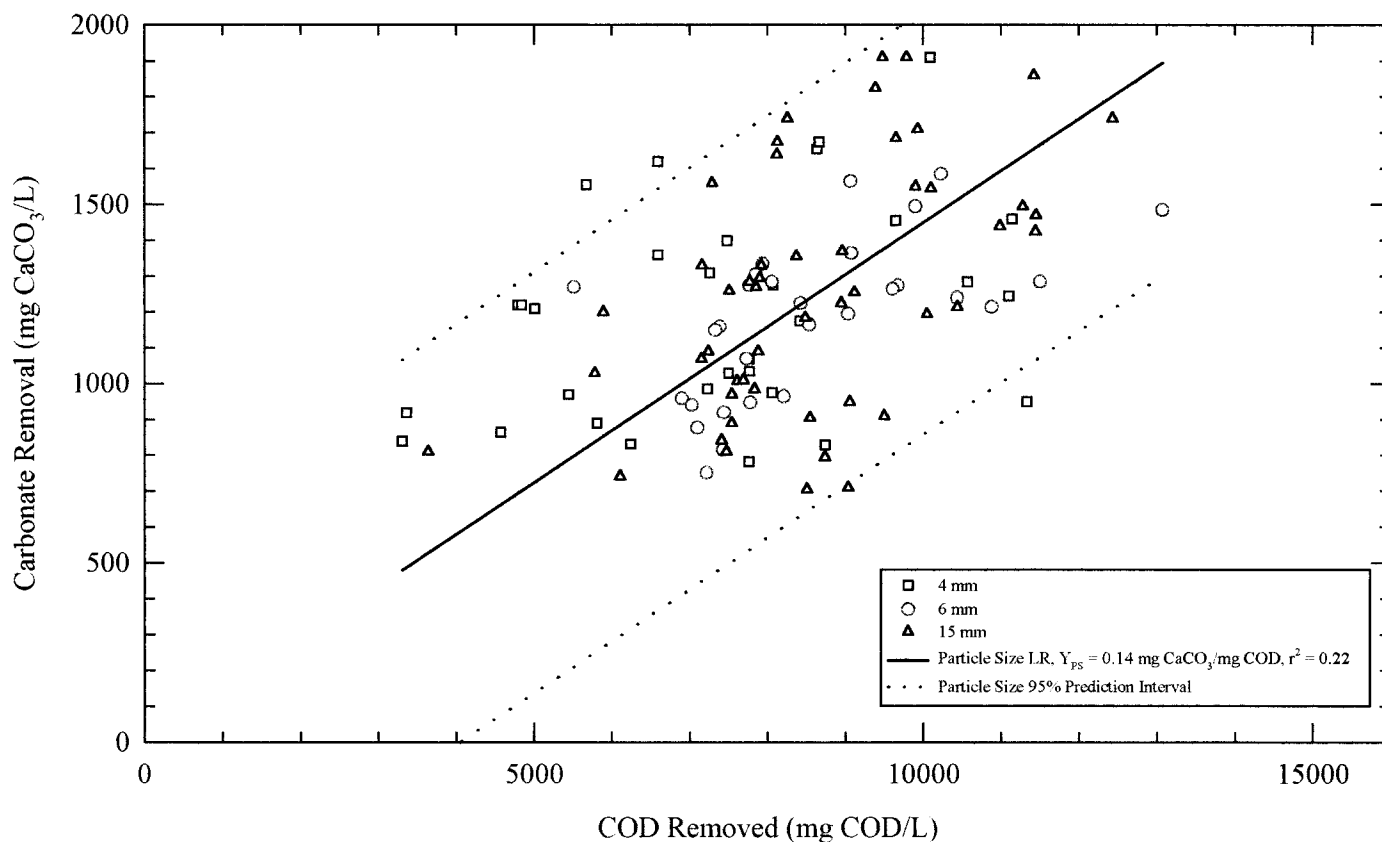


FIG. 4. Measured Calcium Carbonate Removed versus COD Removed and Calculation of Overall Calcium Carbonate Yield Coefficient Y_{PS} by Linear Regression for Columns with Varying Bead Diameters Permeated with Keele Valley Leachate

effect of particle diameter on the microbial population and composition.

The community structure within the columns was examined after column disassembly. The BIOGAS detector indicated the presence of methane-producing bacteria at all levels within the columns. However, there was a clearly defined division between the very high aggressivity (delay times of only 2–3 days) in the lower 200–300 mm above the base (where the leachate entered the column) and the low aggressivity in the upper portion of each column (typical delay times of 8–11 days) for the columns with 4- and 6-mm beads. The larger particle-size columns consistently had a more uniform high aggressivity throughout the column (delay times of 1–3 days) than the columns packed with smaller particles. The BART delay time is a measure of the aggressivity of the microbial population; thus, a short delay time of 2–3 days (1×10^5 to 6×10^5 cells/mL) in the lower portions of the 4- and 6-mm columns and 1–3 days (1×10^5 to 1×10^6 cells/mL) throughout the 15-mm columns implies a large, active population of methanogens within these zones. The long delay times (8–11 days; 5×10^2 – 10^3 cells/mL) in the upper portion (400–600 mm) of the 4- and 6-mm columns implies a low or dormant population of methanogens in this region.

The columns were dominated (Armstrong 1998) by facultative anaerobic bacteria and enterics (e.g., coliforms), yet there were some zones of iron-reducing bacteria present. There was a fairly consistent distribution of dense slime formers and facultative anaerobic bacteria throughout each column, and DN were present at all levels except at the 400-mm level in columns PK3 (6-mm beads) and PK6 (15-mm beads). Finally, there was a highly aggressive population of anaerobic sulfate reducers present throughout the columns.

It is evident from the BART results taken from influent, effluent, and column autopsy samples that there is a large microbial consortia, consisting of methanogens, coliforms, SRB,

DN, and facultative anaerobes present in the column. The presence of these groups of bacteria in landfill waste and leachate has been documented by numerous researchers (Jones et al. 1983; Brune et al. 1994; Palmisano and Barlaz 1995). It is hypothesized that bacteria growing within the decomposing waste detach from the developing biofilms, flow with the leachate into the LCS, and colonize the granular drainage material. The presence of these groups of bacteria signifies that the columns are being colonized by bacteria that would be found in a “typical” landfill, thus the columns are likely simulating the conditions found in a landfill LCS. The increase in effluent leachate sulfate-reducing (SRB) aggressivity correlates with the decrease in aggressivity observed in the SLYM and IRB BARTs. The presence of a large population of methanogens, which thrive in anaerobic environments (Palmisano and Barlaz 1995), in each column indicates that the bacteria not only colonized the columns, but developed niches where specialized bacteria thrive.

DRAINABLE POROSITY

The initial porosity of granular drainage material used in LCSs may range from about 0.3 for sand to about 0.5 for coarse (38 or 50 mm) uniformly graded gravel. To the extent practical, these columns were packed to fall within this continuum with initial porosities of 0.37 and 0.38 for the smaller particles (4 and 6 mm—approximately an orthorhombic packing) and with an initial porosity of 0.5 for the larger particles (15 mm—approximately a cubic packing). Drainable porosity profiles after permeation with leachate are shown in Fig. 5 for the columns packed with 4-, 6-, and 15-mm diameter beads. Fig. 5(a) shows the drainable porosity profiles during steady-state operation when the rate of removal of COD and CaCO₃ is relatively stable (Fig. 3) after 165–171 days. Once the drainable porosity dropped to about 5% or below, it became

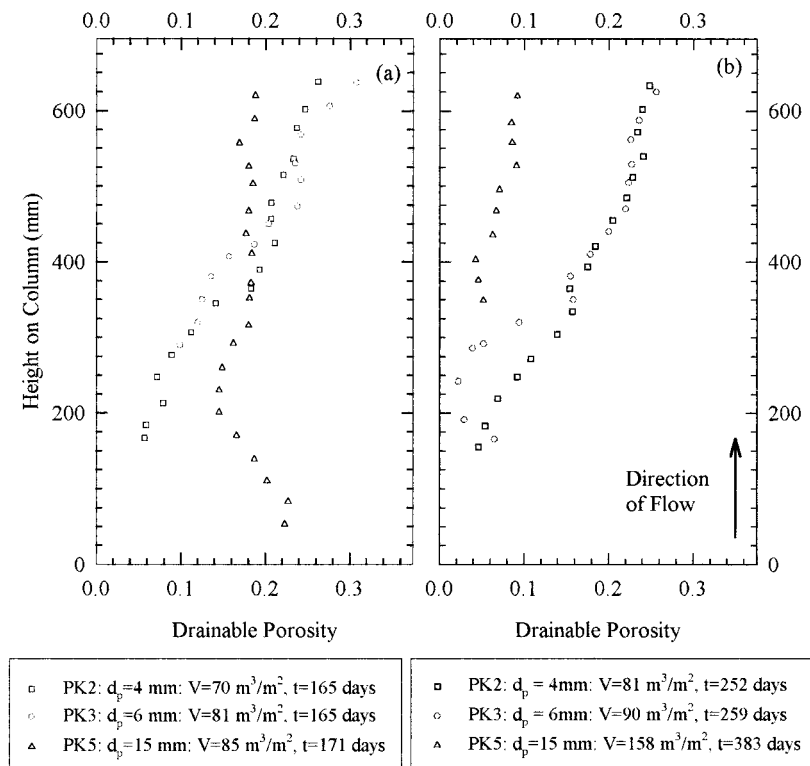


FIG. 5. Drainable Porosity Profiles: (a) after Approximately 165–171 Days; (b) after Column Failure

impractical to measure the drainable porosity in a reasonable period of time because the hydraulic conductivity had dropped to below 10^{-8} m/s (as will be discussed below).

The absence of data points in the lower portion of the columns for the 4- and 6-mm beads is indicative of severe clogging because these are the zones where the column was so clogged the drainable porosity could not be measured (but was below 5%). Fig. 5(a) shows that the clogging is most severe (>86% loss in drainable porosity) at the influent end of the columns for the smaller particles (4- and 6-mm beads) and only limited clogging (<30% drop in drainable porosity) has occurred near the effluent end. In contrast, for the 15-mm beads the drop in porosity was far more uniform (about 60% decrease in drainable porosity) along the entire column.

This is attributed to the combined effect of a number of factors. First, the large surface area for the smaller beads allows more biofilm growth near the influent port for the 4- and 6-mm beads than for the 15-mm beads during the early stages of the tests. This tends to accelerate the clogging in the lower portion of the column. Second, the larger pore size of the 15-mm beads means that, for a given growth of biofilm and mineral clog, there is still a more permeable system to allow leachate through to the upper portions of the columns. Thus for the column with 4- and 6-mm beads, clogging in the lower column begins to reduce the nutrients and CaCO_3 available to the upper portion of the column. With a large particle size, the leachate is more evenly distributed through the column and the clogging is more uniform.

Eventually, significant clogging at the influent end of the column made it impractical to maintain the design flow rate. [Plots of flow versus time are given in Fig. 2]. The column was defined as having “clogged” when it was not possible to transmit >10% of the daily design flow rate under a head of 2.4 m. Drainable porosity profiles for the 4-, 6-, and 15-mm bead diameter columns are shown in Fig. 5(b), just prior to termination of the tests. At failure, the 4-, and 6-mm columns have similar drainable porosity profiles, which vary from <5% below 300 mm above the column base to 25% at 600 mm

above the column base. The 15-mm bead column had a more consistent drainable porosity of between <5 and 9%. The profiles in Fig. 5(b) indicate that the 4- and 6-mm bead columns failed due to accumulation of clog material in the lower half of each column. The 15-mm bead column appears to have failed due to a more consistent accumulation of clog material along the entire column length.

The 15-mm bead column had a greater absolute and percentage decrease in drainable porosity, but this also occurred over a much longer period of time. Operation of the 15-mm bead column PK5 was discontinued after 383 days of operation, but the actual flow rate decrease, from design flow rates, began after 329 days with $152 \text{ m}^3/\text{m}^2$ of leachate passed (Fig. 2). The 6-mm column PK3 was discontinued after 259 days, but the actual flow rate decrease, from the design flow rate, began after 140 days and $75 \text{ m}^3/\text{m}^2$ of leachate. Finally, the 4-mm PK2 column was discontinued after 252 days, but the actual flow rate decrease began after 132 days with $68 \text{ m}^3/\text{m}^2$ of leachate. Thus the effective service life (the period of time during which the design flow could be maintained) of the 15-mm beads (329 days) was more than twice that of the 6-mm (140 days) and 4-mm (132 days) beads.

Cumulative CaCO_3 calculations show that, relative to the total packed volume of the columns, the 15-mm bead column accumulated approximately $0.23 \text{ g}/\text{cm}^3$ of CaCO_3 compared to $0.12 \text{ g}/\text{cm}^3$ and $0.09 \text{ g}/\text{cm}^3$ for the 6- and 4-mm beads, respectively. The 15-mm bead column has an average of 0.88 g CaCO_3 removed per liter of leachate through the column compared to 0.82 and 0.69 g CaCO_3 removed per liter for the 6- and 4-mm-bead columns, respectively. This suggests that the 15-mm-bead column has an increased removal efficiency, when compared to the 6- and 4-mm-bead columns, while operating over a longer period of time and allowing passage of a larger volume of leachate. This is attributed primarily to the fact that the pore spaces between the larger particles in the 15-mm-bead column tend to remain free of obstructions for a greater period of time than the 6- and 4-mm columns (e.g., Fig. 5).

HYDRAULIC CONDUCTIVITY

The initial hydraulic conductivities k_{init} were measured using the constant-head method (ASTM D 2434-68). For a given column, the hydraulic conductivities k at various times were then normalized by dividing these values by the initial value for that column ($k_{init} = 0.16, 0.33,$ and 1.4 m/s for 4-, 6-, and 15-mm beads, respectively). Thus a value of normalized hydraulic conductivity k/k_{init} of 1 implies no change in hydraulic conductivity. A value of $k/k_{init} = 10^{-7}$ implies 7 orders of magnitude drop in hydraulic conductivity from the initial value.

The variation in normalized hydraulic conductivity k/k_{init} between the first two piezometers (between 26 and 140 mm from the inlet end of the column) with cumulative flow [Fig. 6(a)] and elapsed time [Fig. 6(b)] are shown in Fig. 6. Because there were two 4-mm columns, two 6-mm columns, and two 15-mm columns, two data points often are shown at a given time for a given bead size. The difference between these data points reflects the different responses of the two different columns and hence is a measure of variability. However, in general terms, the results for a given particle size were very consistent.

As previously noted in the methodology section, it was not practical to collect data until the hydraulic conductivity had dropped by 5 orders of magnitude. Thus, for the 4-, and 6-mm beads, the head difference between piezometers P1 and P2 was not measurable and, hence, there are no data for the first 95 days of testing. For the 15-mm beads, it took 320 days for the hydraulic conductivity to drop to the point where there was a measurable head difference between piezometers P1 and P2. Referring to Fig. 6(b), it can be seen that after 95 days

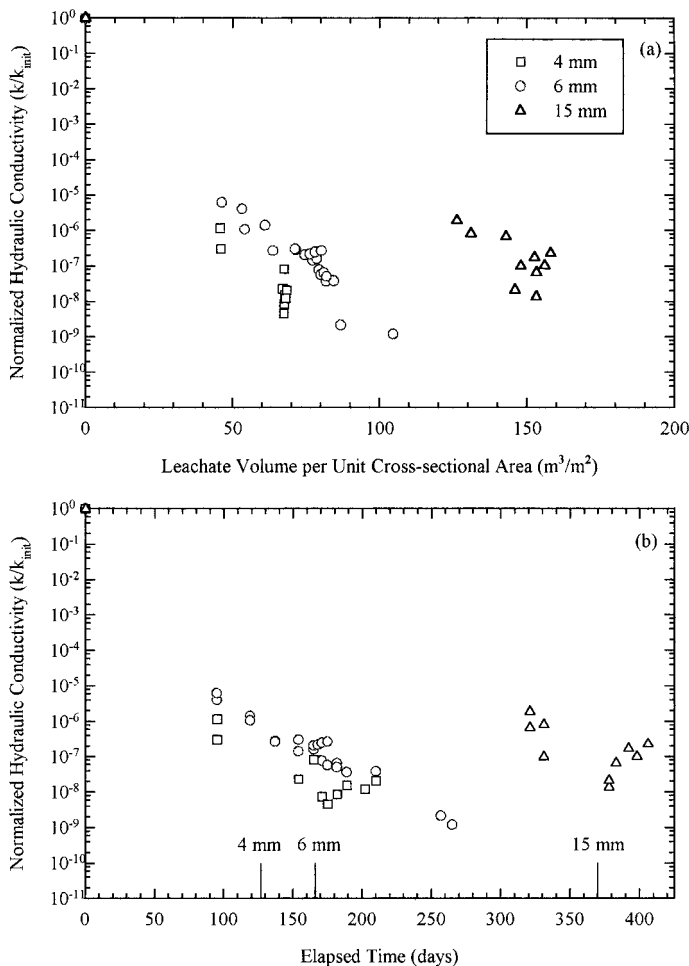


FIG. 6. Hydraulic Conductivity versus: (a) Leachate Volume per Unit Cross-Sectional Area; (b) Elapsed Time

the drop in hydraulic conductivity was consistently greater for the two columns with 4 mm than for the two columns with 6 mm. If one were to draw a horizontal line corresponding to a 6 orders of magnitude drop in hydraulic conductivity [$k/k_{init} = 10^{-6}$ on Fig. 6(b)], one will see that it took about 95 days for the 4-mm beads, 120 days for the 6-mm beads, and 320 days for the 15-mm beads to experience a 6 orders of magnitude drop in hydraulic conductivity between piezometers P1 and P2.

Every effort was made to maintain a constant flow rate Q_d during the tests; however, as the hydraulic conductivity between piezometers P1 and P2 dropped, the head in piezometer P1 increased (for a given flow rate). There were practical limits to the head that could be permitted in P1 without leachate overflowing into the gas collection bag (Fig. 1). Due to this limit, the flow could not be maintained at the specified rate once the hydraulic conductivity had dropped by about 7 orders of magnitude [$k/k_{init} \approx 10^{-7}$ in Fig. 6(b)] and the flow rate had to be reduced. This occurred after 127, 166, and 370 days for the 4-, 6-, and 15-mm-bead columns, respectively [shown by the short vertical lines at the bottom of Fig. 6(b)].

Referring to Fig. 6(a), it can be seen that a 6 orders of magnitude drop in hydraulic conductivity (i.e., $k/k_{init} = 10^{-6}$) corresponded to about $45 \text{ m}^3/\text{m}^2$ of leachate through the 4-mm beds compared to about $63 \text{ m}^3/\text{m}^2$ for 6-mm beads and $130 \text{ m}^3/\text{m}^2$ for 15-mm beads. Thus, for equal losses of hydraulic conductivity, the 4-mm-bead columns passed the smallest volume of leachate. This is because the 4-mm beads had the smallest voids. Therefore, for a similar film growth on the beads, the pore space became constricted for the 4-mm beads prior to the pore space being constricted for the 6-, and 15-mm beads.

As early as 1922, Blake (1922), and subsequently Kozeny (1927), Carman (1937, 1956), and many others, attempted to develop theoretical and empirical relationships between porosity and hydraulic conductivity. However, these relationships have been developed for "clean" porous media. Although they may start clean, LCSs, and the columns examined herein, are not clean systems. They start with an initial porosity that decreases with time due to development of biofilm and, most significantly, an inorganic deposit on the original particles (to be discussed in more detail in the next section). This leads to a decrease in hydraulic conductivity with drainable porosity. To the writers' knowledge, no results have been published previously for the case of uniform porous media that has experienced a drop in drainable porosity and hydraulic conductivity due to a buildup of encrustation in the system caused by biodegradation of landfill leachate as it passes through the porous media.

Models are presently being developed to predict the rate of clogging and the service life of different LCSs. These models (Rowe et al. 1997; Cooke et al. 1999) can relate the leachate loading and time to the buildup of both biofilm and inorganic precipitates and hence can be used to calculate a decrease in porosity with time. However, to convert this to a change in hydraulic conductivity, it is necessary to have a relationship between hydraulic conductivity and porosity that has been developed for deposition conditions similar to that expected in LCSs. Thus an attempt was made to identify such a relationship from the porosity and hydraulic conductivity measurements conducted between piezometers at different times in this test program. It was found that the following empirical relationship between hydraulic conductivity k and drainable porosity n provided a reasonable fit to the data for the 4-, 6-, and 15-mm glass beads ($r^2 = 0.94$; Fig. 7):

$$k = 1.17 \times 10^{-9} \exp(49.85n) \text{ (m/s)}, \quad n \geq 0.03 \quad (1)$$

It can be seen that the hydraulic conductivity drops with decreasing drainable porosity in a similar manner for all par-

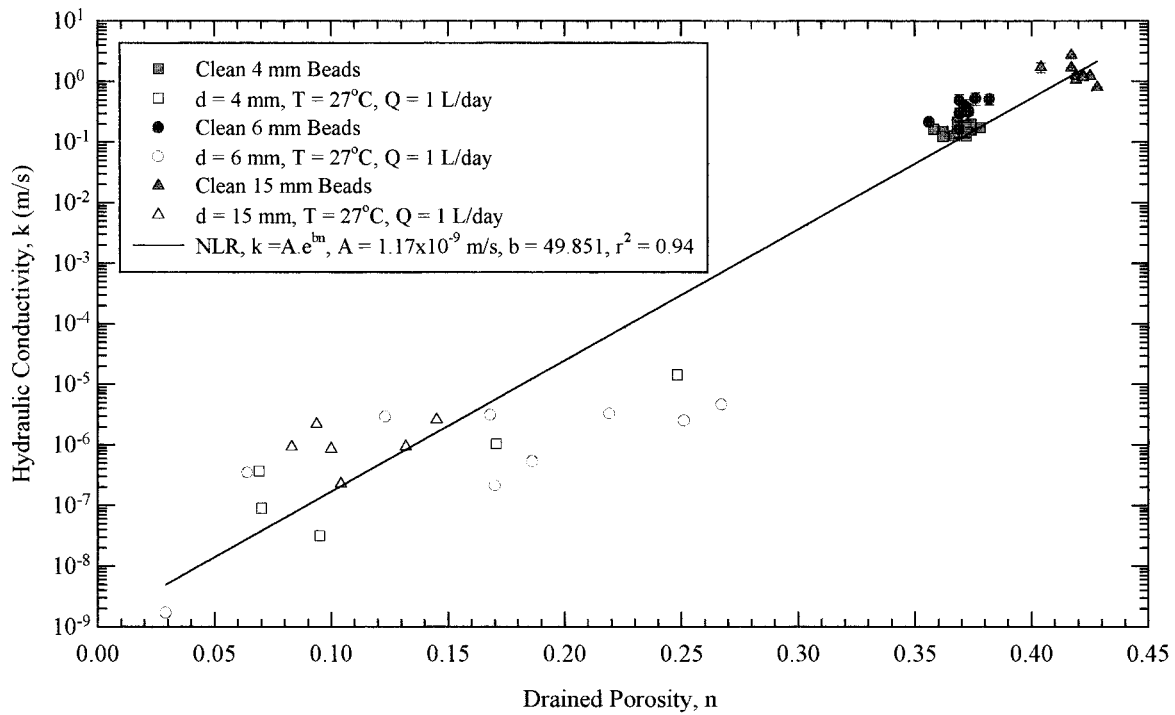


FIG. 7. Relationship between Drainable Porosity and Hydraulic Conductivity with Respect to Particle Diameter

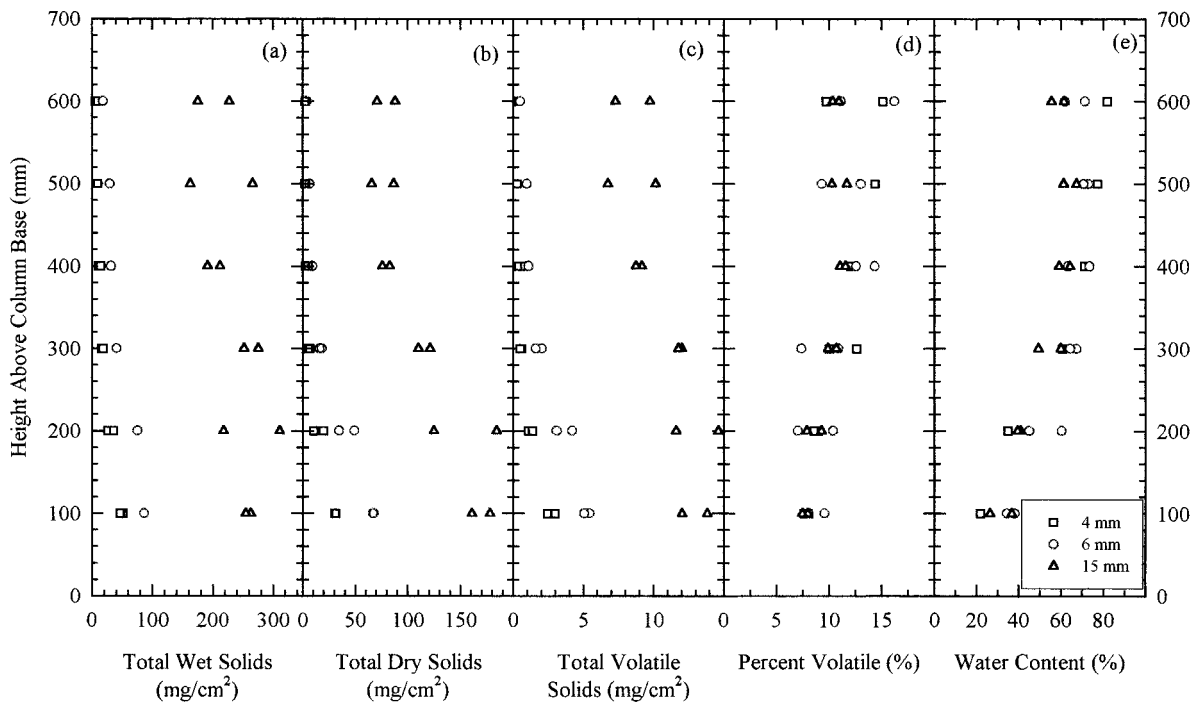


FIG. 8. Characteristics of Clog Material at Termination of Tests: (a) Total Wet Solids Mass per Bead Normalized by Bead Surface Area; (b) TDS Mass per Bead Normalized by Bead Surface Area; (c) TVS Mass per Bead Normalized by Bead Surface Area; (d) Percent Volatile; (e) Water Content

ticle diameters and that there is about a 7–8 orders of magnitude drop in hydraulic conductivity by the time the drainable porosity drops to about 10% of the initial value.

COLUMN DISASSEMBLY

The columns were disassembled at the conclusion of the operational phase and the distribution and characteristics of the clog material were examined. Fig. 8 shows the distribution along the column of total wet solids, total dry solids (TDS), and total volatile solids (TVS) as mass per unit surface area

of bead. For these parameters, the three particle diameters show the same trend of decreasing mass per bead with increasing distance from the column base, where the leachate is located. This is consistent with the trend in drainable porosity that is least near the column inlet and increases as one moves up the column [Fig. 5 (b)]. However, it also was evident that there is generally more clog material per unit surface area of bead along the columns for the larger particle diameter (15 mm) than for the smaller particle sizes (6 and 4 mm) over the entire length of the column. However, although the percentage

of volatile solids increases as one moves up the column, there is no evident trend due to the particle diameter in terms of the proportion of volatile solids. The larger particle diameter has larger pore spaces for the accumulation of clog material along the entire length of the column (giving high TDS at all elevations) by the time the tests were terminated. This appears to have resulted in more active (TVS) and inactive (TDS) clog material, although the ratio of active biofilm to inactive encrustation (expressed in terms of the percentage of volatile solids) does not seem to change significantly with particle diameter. When considering the total distribution of clog material per unit length of column, it is important to note that the number of beads per unit volume varies from an average of about 18.8 beads/cm³ for 4-mm beads to 5.5 beads/cm³ for 6-mm beads and 0.28 beads/cm³ for 15-mm beads. The specific surface (surface area per unit volume) is about 9.4, 6.2, and 2 cm²/cm³ for 4-, 6-, and 15-mm beads, respectively.

The bulk biofilm density, biofilm thickness, and void volume occupancy were calculated for samples taken during the disassembly procedure. The bulk density of clog was between 1.64 and 1.78 g/cm³, with an average of 1.73 g/cm³ as evaluated using ASTM Method D 854. In discussing the distribution of clog material, it is useful to recall that the 4-mm columns clogged first and hence were run for a shorter period of time than the 6- and 15-mm columns. Thus the mass loading prior to termination increased with particle size, because it took longer for the columns with larger particles to clog (i.e., experience the same drop in hydraulic conductivity and drainable porosity between piezometers P1 and P2).

It also should be remembered that the void size is related to particle size. Adopting the relationship by Kennedy et al. (1985) that the characterization opening size is 0.2 D₁₅, it follows that the characteristic opening sizes were 0.8 mm for 4-

mm beads, 1.2 mm for 6-mm beads, and 3 mm for 15-mm beads. Thus, for a given buildup of active (biofilm) and inactive clog material on a bead, the available opening size should be greater for larger particles. For example, the buildup of a 0.2-mm-thick film on the three different bead sizes would represent 25, 17, and 6.7% of the characteristic opening size for 4-, 6-, and 15-mm beads, respectively. Thus it is not surprising that for the same initial mass loading, by the time the 4-mm-bead column had clogged, the 6- and 15-mm-bead columns were not yet clogged, as previously discussed. Likewise, it is not surprising that the clog thickness at 100 mm above the column base (calculated from the average total clog wet mass and the clog bulk density) increased with increasing particle diameter.

Samples were taken from the 25–75 mm position for columns PK2 (4-mm beads), PK3 (6-mm beads), and PK5 (15-mm beads) to compare clog composition with respect to particle diameter. The analysis program inductively coupled plasma spectrophotometry (EPA Method No. 6010) for trace metals, alkaline metals, and sulfur using a Thermo Jarrell Ash ICAP 61E Plasma Spectrophotometer. Silicon was analyzed by inductively coupled plasma-atomic emission spectroscopy and conversion to silica [ASTM Standard Methods (1985) 425A (modification)]. Total Kjeldahl nitrogen (TKN) was analyzed by continuous liquid flow (ASTM Method D3590-84A&D). Volatile solids were obtained by ignition to constant weight at 600°C (EPA Method No. 160.4).

The results from the elemental analysis are summarized in Table 1. Although there is some scatter in the data, there is no evidence of the effect of particle diameter or the corresponding different lengths of the tests on calcium or carbonate content. The average calcium content of 25.7% was also similar to the value of 25% obtained in the field by Fleming et al. (1999).

TABLE 1. Elemental Analysis of Clog Material (25–75 mm Position)

Parameter (1)	4 mm (2)	6 mm (3)	15 mm (4)	Average (5)
Water content (% wet)	44.9	41.2	22.1	36.1
TOC as C (% dry)	3.5	6.7	5.6	5.3
Organic matter (TVS) (% dry)	12.4	11.2	9.7	11.1
Carbonate as CO ₃ (% dry)	49.4	49.8	51.6	50.3
Ca (% dry)	26.8	24.2	26.2	25.7
Si (mg/kg)	39,700	33,900	19,100	30,900
Mg (mg/kg)	14,400	12,700	5,870	10,990
Na (mg/kg)	2,570	3,060	2,000	2,543
Fe (mg/kg)	30,700	37,000	26,800	31,500
Al (mg/kg)	141	163	98	134
K (mg/kg)	1,160	1,800	889	1,283
S (mg/kg)	6,280	10,100	3,880	6,753
Total P (mg/kg)	1,940	2,140	1,560	1,880
TKN-N (mg/kg)	9,400	10,000	6,500	8,633
Mn (mg/kg)	1,890	1,720	1,840	1,817
Ti (mg/kg)	21	27	22	23
Zn (mg/kg)	1,100	1,500	419	1,006
Sr (mg/kg)	818	751	966	845
Ba (mg/kg)	102	88	85	92
Ni (mg/kg)	78	108	47	78
Cu (mg/kg)	134	16	7	52
V (mg/kg)	15	17	15	16
Pb (mg/kg)	<1	<1	<1	<1
Cr (mg/kg)	13	14	16	14
Co (mg/kg)	13	12	4	10
B (mg/kg)	15	15	10	13
Ca/CO ₃ ^a	0.54	0.49	0.51	0.51
Mg/TVS ^a	0.12	0.11	0.06	0.10
Na/TVS ^a	0.021	0.027	0.021	0.023
Fe/TVS ^a	0.25	0.33	0.28	0.28
S/TVS ^a	0.051	0.090	0.040	0.061
TKN/TVS ^a	0.076	0.089	0.067	0.078
Zn/TVS ^a	0.009	0.013	0.004	0.009

^aDimensionless mass ratio.

TABLE 2. Comparison of Keele Valley Leachate Composition with Clog Material Composition

Parameter (1)	Keele Valley leachate geometric mean 1997 (mg/L) (2)	Clog material average (mg/kg) (3)	Ratio (-) (4)
DOC/TOC	4,184	52,600	13
Ca	536	257,000	480
Mg	401	10,990	27
Na	1,864	2,543	1.4
Fe	205	31,500	154
Al	0.94	134	143
K	856	1,283	1.5
S ^a	46	6,753	147
tot P	0.72	1,880	2,611
TKN-N	1,065	8,633	8.1
Mn	6.30	1,817	288
Ti	0.12	23	202
Zn	5.67	1,006	177
Ba	0.29	92	321
Ni	0.65	78	119
Cu	0.01	52	4,223
V	0.03	16	614
Cr	0.15	14	93
Co	0.03	10	324

^aSO₄ in leachate converted to S.

The silicon may be contaminated from the cutting process where clog material and glass beads were cut to remove samples.

The average results from the elemental analysis on these columns are summarized along with the average Keele Valley leachate parameters in Table 2 for all parameters where data are available both in the leachate and clog material. The ratio of concentration in the leachate to that in the solid clog material indicates that parameters, except Na and K, tend to accumulate in the solid clog to some degree. Calcium shows one of the highest accumulation ratios, and this reflects the significant precipitation of CaCO₃ that has occurred as the leachate passed through the columns. Magnesium has a greater solubility than calcium, and it does not accumulate as rapidly, despite similar concentrations in the leachate. Na and K show no accumulation. Total organic carbon (TOC) and TKN have accumulation ratios that are close and small relative to most other parameters examined. Because TKN represents the nitrogen in organic matter, it is likely that the accumulation of TKN and at least a portion of the TOC and P, are related to biofilm growth and not precipitation. The accumulation rate for phosphorus is about 3 orders of magnitude greater than other parameters required for cellular growth, but the leachate concentration is between 3 and 4 orders of magnitude smaller than these other parameters. In the clog material, the ratio of TOC:N:P is 28:4.6:1. It has been noted that C:N ratios for heterotrophic bacteria are difficult to quantify because carbon is used for catabolic (cell growth) and synthesis (e.g., extra polymer substances) functions (Cullimore 1993). The N:P ratio (typically 4:1–8:1) is suggested to be a better measure because these elements are essentially retained within the cellular structure, with the N:P ratio for the clog material of 4.6:1. This suggests that TOC, N, and P accumulations are related to the biofilm growth. The sulfur accumulation ratio is higher than the other listed parameters, because it also accumulates as FeS (Brune et al. 1994). Other parameters in order of decreasing accumulation ratio are Cu, V, Co, Ba, Mn, Ti, Zn, Fe, S, Al, Ni, and Cr.

FIELD COMPARISON

Because both the first and the second Toronto landfills had the same waste stream, it is assumed that the leachate strength

in the early life of the second landfill was similar to that at the first landfill (Keele Valley) that was used for the column tests. Thus, it is hypothesized that the columns can be used to estimate the time required for the pea gravel around the leachate collection drain at the second landfill to fail due to clogging (i.e., experience a reduction in hydraulic conductivity of about 7–8 orders of magnitude).

Although pea gravel was used in the landfill French drain and glass beads were used in the present study, the effect of the surface chemistry of the drainage material was not considered to be a major factor because this surface was quickly covered with biofilm (within 20 days in the present experiments) and the surface reactions were then controlled by the biofilm surface. As clogging progresses, a buildup of material with a composition as given in Table 1 occurs. A related concern has been that leachate with a pH < 7 might attack the drainage material if dolomitic limestone is used for the granular material. However, Bennett et al. (2000) have shown that dolomitic limestone does not experience significant dissolution in landfill LCSs. This is because the leachate is supersaturated with respect to carbonate minerals. A secondary neofomed calcite is quickly deposited on, or envelops, the granular particles, and this occurs for both glass beads and crushed limestone.

As previously discussed, column flow rate was selected to simulate the estimated horizontal Darcy flux through the saturated zone adjacent to the collection pipe at this landfill. At this landfill, leachate flows through the waste until it reaches a French drain that has a radius of about 0.5 m. This represents a change in conditions and clogging of the pea gravel can be expected to occur as the leachate flows from the waste into the pea gravel. Considering the flow into one half of the pipe (i.e., assuming a line of symmetry about the vertical axis of the pipe), the surface area A_f of gravel through which leachate has the potential to progressively flow (as the clogging proceeds from bottom up) is given by

$$A_f = 0.5 \times \pi \times r \times w_e \quad (2)$$

where r = radius of stone; and w_e = length along the pipe being considered (e.g., the longitudinal spacing between perforations). Based on the columns test, it is known that clogging was deemed to have occurred (due to inability to maintain a flow exceeding 10% of the design flow) for average flow per unit area V_N of 68.2, 98.8, and 161 m³/m² for particle sizes of 4, 6, and 15 mm, respectively. As previously noted by Koerner et al. (1994), rate of clogging can be related to the mass loading per unit area. In the present context, this corresponds to the flow per unit area. Hence, under similar flow conditions, one would expect clogging of the pea gravel at this landfill to occur when the ratio of total flow to the area through which this flow enters the gravel was similar to that in the columns

$$\frac{Q_f \times t}{A_f} = V_N \quad (3)$$

where Q_f = flow to the French drain and is given by

$$Q_f = q_0 \times L \times w_e \quad (4)$$

Solving for time t and rearranging gives

$$t = \frac{0.5 \times \pi \times r}{q_0 \times L} \times V_N \quad (5)$$

Based on the average volume of fluid per unit area required to cause clogging, as noted above, and taking $r = 0.5$ m, one can then calculate the time for clogging assuming a percolation $q_0 = 0.1$ m/a (typical of the area, where a = annum), as summarized in Table 3. It can be seen that the estimated time to clog is between 5 and 13 years. Because it was observed that a leachate mound had developed after 11 years of landfill op-

TABLE 3. Calculated Time for Clogging of the French Drains at Landfill 2 ($L = 100$ m, $r = 0.5$ m)

Particle diameter, dp (mm) (1)	Estimated saturated thickness, h (m) (2)	Percolation rate, q_0 (m/a) (3)	Observed volume of fluid to clog, V_N (m^3/m^2) (4)	Calculated time to clog, t (years) (5)
4	0.05	0.1	68.2	5.3
6	0.05	0.1	98.8	7.8
15	0.05	0.1	161	12.6

erations, it appears that the rate of clogging one would predict based on these tests is generally consistent with what was observed.

CONCLUSIONS

The results from this study have shown that particle diameter has a significant impact on the rate and extent of clogging in a granular media. This is primarily due to the increased pore size associated with larger characteristic particle size that increase the time required for occlusion of pore spaces. The blockage of pores by accumulation of organic and, in particular, inorganic clog material reduces the hydraulic conductivity of the porous media and, in a field case, would reduce the effectiveness of the drainage layer. It is shown that clogging is greatest where there is greatest mass loading (near the inlet in this case, but likely near the collection pipes in a field situation).

It was found that larger particle diameters gave rise to bio-reactors as efficient as smaller particle diameters (i.e., a similar reduction in organic and inorganic loading in a given time). This appears to have been because the clogging focused over a similar surface area—near the inlet of the columns for the smaller particles that had a large surface area per unit volume and over the entire column for the larger particles. This is evident from the drainable porosity profiles that show that leachate flowing through the smaller particle sizes (4 and 6 mm) quickly reduces the porosity in the lower third of each column to the point where the hydraulic conductivity drops by 7 to 8 orders of magnitude and the flow drops to below 10% of the design flow rate, whereas the 15-mm-bead columns showed a much more consistent reduction in drainable porosity throughout the column over time, reaching a similar reduction in drainable porosity throughout the column at failure. The surface area of beads in the first 130 and 200 mm of the 4- and 6-mm-bead columns, respectively, is approximately equal to the total surface area in the 620-mm packed length of each 15-mm column.

The microbiological testing indicated that the microbial consortia found within the columns consisted of methanogens, denitrifiers, sulfate reducing, and other facultative anaerobes. The results suggest that the columns simulate the microbiology that has been found in landfills. The distribution of the methanogenic bacteria within the columns suggests that the very high aggressivity found within the lower 300 mm of the column with 4- and 6-mm beads and throughout the 15-mm-bead columns may be related to the most intensely clogged zones within the columns.

The results of this study lead to several conclusions of practical significance concerning the design and operation of an LCS. First, increasing the diameter of the granular media (D_{10}) used in an LCS may increase the LCS operational life span, with all other factors being equal. Second, this study has found bulk densities of clog material ρ_c between 1.6 and 1.8 Mg/m³. This combined with the observed total calcium fraction of 26%

of total clog material ($f_{ca} \approx 0.26$) can be used in simple engineering calculations such as those proposed by Rowe and Fleming (1998) to estimate the rate of clogging of different collection system designs. The yield coefficient Y_c of CaCO₃ did not vary significantly with particle diameter. This is an important parameter needed in more sophisticated models of the rate of clogging of LCSs (Rittmann et al. 1996; Rowe et al. 1997; Cooke et al. 1999).

Finally, it was found that the time to clogging of a field case was reasonably predicted using the results obtained from the laboratory tests, suggesting that the tests reasonably simulate what is occurring in the field.

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