Modelling species fate and porous media effects for landfill leachate flow

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Abstract: A numerical, multiple-species, reactive transport model, coupled to models of kinetic biodegradation, precipitation, and particle attachment and detachment for predicting landfill leachate-induced clogging in porous media for one-dimensional flow systems, is described. The finite-element method is used for transport modelling, with reactions incorporated into point-source or sink terms. The species modelled include three volatile fatty acids, active and inert suspended biomass, dissolved calcium, and inorganic particles. The clog matter consists of active biofilm, inert biofilm, and inorganic solids. A biofilm model is used to simulate the growth and decay of active biomass and removal of substrate. Precipitate accumulation and calcium removal are simulated by a model of calcium carbonate precipitation. Interphase movement between clog matter and fluid includes the processes of attachment and detachment. A geometric representation of the porous media allows porosity and specific surface to be estimated from the thickness of the accumulated clog matter. The porosity of the media can thus change spatially and temporally. The behaviour of the model is demonstrated with a hypothetical example.

Key words: clogging, landfills, leachate collection systems, modelling, biofilms, mineral precipitation.


Mots clés: colmatage, remblais, systèmes de collecte de lixiviant, modélisation, biofilm, précipitation minérale.

Introduction

Modern solid-waste landfills are designed with a barrier system beneath the waste material, to control the migration of harmful fluids (leachate) and minimize contamination of the environment. The barrier system often includes a leachate collection system (LCS), which is a high-permeability drainage layer (or layers) designed to control the height of leachate mounding on the landfill base (and thus minimize advective flow of leachate from the system) and to allow removal of leachate for treatment or recirculation. The LCS is typically a layer of granular material, such as sand, gravel, or crushed stone, graded to slope to perforated pipes. For the LCS to function adequately, its hydraulic conductivity must be sufficiently high to maintain the leachate head on the landfill base at or below the design hydraulic head. However, the hydraulic conductivity of the LCS in some landfills can decrease significantly as a result of the buildup of biomass, precipitate, and sediment (Bass 1986; Brune et al. 1991; Koerner et al. 1993; McBean et al. 1993; Rowe et al. 1993).
1995; Rowe 1998; Fleming et al. 1999; Maliva et al. 2000; Bouchez et al. 2003). When a collection system can no longer maintain the leachate head below the design level, it is said to have clogged and hence reached its service life. To develop landfill designs that will provide good long-term performance, it is necessary to be able to predict the service life of the LCS. Furthermore, being able to predict the rate of biologically induced clogging may lead to changes in LCS design that will increase the service life.

As part of a decade-long study of the clogging of LCSs, the authors and their collaborators have been developing (i) a numerical model for clogging processes; (ii) experimental data based on controlled column tests (Rowe et al. 2000a, 2000b, 2002; VanGulck and Rowe 2004a, 2004b) to provide insight regarding the mechanisms to be modelled and to verify the model; and (iii) mesocosm (laboratory-cell) tests that simulate, in real time and at a realistic scale, the clogging process in LCSs (Rowe et al. 1995; Fleming et al. 1999; Fleming and Rowe 2003) over a period of up to 10 years.

As a first step toward modelling the clogging of landfill LCSs, the authors developed a model and applied it to column experiments conducted with synthetic leachate (Cooke et al. 1999, 2001). Although this model worked well for the column tests with synthetic leachate, application of the model to columns involving real leachate identified a need to incorporate additional mechanisms to give good predictions of clogging caused by real leachate.

The new model presented here represents a significant advancement over earlier models, in that it predicts the fate of butyrate (as well as of propionate and acetate), dissolved calcium, suspended active biomass, suspended inert biomass, and suspended inorganic solids. The process of attachment is now modelled (for all suspended solids), along with a new inert biofilm layer. The method of predicting precipitation of calcium carbonate (CaCO₃) is also improved. Finally, transport is modelled by using the finite-element method, with species reactions being incorporated in source or sink terms. The current model has been applied by VanGulck (2003) to bead-packed columns permeated with synthetic and real landfill leachate and by Cooke et al. (2005) to gravel-packed columns permeated with real leachate.

The objective of this paper is to describe all significant aspects of the new, generalized, one-dimensional (1D) transport and clogging model and to demonstrate its use for a simple problem. The following sections describe the basic mechanisms incorporated in the clogging model, together with the rationale for their incorporation. The details of the method for incorporating these mechanisms are then presented. Finally, the application of the model is illustrated for a simple scenario of 1D flow.

Solute transport model

The partial differential equation for saturated, non-uniform solute transport in one dimension is

\[ \frac{\partial (nC)}{\partial t} = nD_x \frac{\partial^2 C}{\partial x^2} - \frac{\partial}{\partial x} (v_x C) + S_R \]

where \( t \) is time; \( x \) is a space coordinate; \( n \) is porosity; \( C \) is solute (species) concentration; \( D_x \) is a dispersion coefficient; \( v_x \) is the Darcy flux (Darcy velocity) in the \( x \)-coordinate direction; and \( S_R \) is a source or sink term for the addition or removal of species through a biogeochemical reaction. Techni-ques for solving this partial differential equation using the finite-element method, subject to appropriate initial and boundary conditions, are well developed (e.g., Istok 1989) and have been adopted here. This paper describes the novel features of the proposed model that have been developed to address issues of clogging; these features include the evaluation of the source or sink term \( S_R \) and the calculation of the change in porosity \( n \) with position and time.

For the purposes of modelling clogging in laboratory columns and LCSs, eq. [1] is subject to up to two of the following four boundary conditions: (i) prescribed concentration (Dirichlet); (ii) prescribed dispersive flux (Neumann); (iii) prescribed total flux (Cauchy); and (iv) non-prescribed dispersive flux (open or free). Boundary conditions (i) and (ii) were described by Istok (1989), whereas boundary conditions (iii) and (iv) were derived from Frind (1988). Details are given by Cooke. The finite-element formulation adopted here uses linear elements with two nodes. A relaxation term is used so that forward, central, or backward difference methods may be used to march the solution forward in time from the initial conditions. However, unlike conventional transport modelling, the porosity \( n \) and the source or sink term \( S_R \) vary as the clogging processes develop and must be recalculated at each time step. The mechanisms causing these changes are described in the next section, followed by a description of the techniques for evaluation of these parameters.

Clogging mechanisms modelled

Choices made in the development of this model (referred to as BioClog in the following discussion) have been largely based on experimental evidence elaborated in the subsequent sections. These findings indicate that the following primary clogging mechanisms should be included in the model: (i) accumulation of active and inert biomass; (ii) accumulation of CaCO₃ precipitate; and (iii) accumulation of inorganic particles originally suspended in the leachate. Each of these mechanisms is discussed below.

Accumulation of biomass

Background and rationale

Biomass accumulation is partly responsible for clogging drainage layers in LCSs. Bass (1986) reported several cases of excessive biological clogging of drainage layers and concluded that the conditions for this process are present in municipal solid-waste landfills. In a survey of landfills in Germany, Brune et al. (1991) found a significant amount of organic material mixed with the consolidated, insoluble deposits in the drainage layers. Likewise, Koerner et al. (1993) reported finding agglomerations of fines and biomass filling the void spaces of poorly graded crushed stone in a landfill
drainage trench. McBean et al. (1993) and Rowe (1998) cited examples of landfills in which the accumulation of biomass caused mounding and consequently leachate seeps, and Fleming et al. (1999) exhumed a landfill drainage blanket after only 4 years of exposure to leachate and found significant quantities of soft black biomass coating the stone and occupying much of the void space.

Column experiments with real or synthetic landfill leachate, performed to observe clogging, have shown that the anaerobic bacteria in the leachate easily colonize the surfaces of drainage materials and form a thick biofilm (Brune et al. 1991; Pakry et al. 1995, 1998; Peeling et al. 1999; Rowe et al. 2000a, 2000b, 2002; VanGulck and Rowe 2004a, 2004b). Rowe et al. (2000a, 2000b) found that the bacteria included methanogens, denitrifying bacteria, and sulfate-reducing bacteria. Pakry et al. (1995) concluded that the bacterial populations remove volatile fatty acids (VFAs) from the leachate, and the growth of these bacteria results in a decrease in drainable pore volume of the drainage material. The biomass can readily adapt to changes in leachate composition and organic loading (Peeling et al. 1999). Brune et al. (1991) and Rittmann et al. (1996) also indicated that the microbial activity of bacteria mediates the formation of incrustations in drainage systems.

High concentrations of suspended bacteria (sometimes measured as volatile suspended solids (VSS)) have been found in landfill leachate and in the influent and effluent of column experiments simulating leachate clogging (Brune et al. 1991; Rowe et al. 2000a, 2000b; VanGulck and Rowe 2004a). In columns containing 6 mm glass beads and permeated with landfill leachate, VanGulck and Rowe (2004a) found that 60%–95% of the influent VSS was removed (by attachment). By comparing results from column experiments using leachate collected from a landfill (referred to here as real leachate) and synthetic leachate (produced in the laboratory to have a similar chemical composition as real leachate but without significant VSS), Rowe et al. (2002) also found that the greater quantity of VSS in real leachate increases the rate of clogging relative to synthetic leachate.

Active biomass experiences endogenous decay, where some cells self-oxidize to gain energy and electrons for maintenance of other cells. The decayed cells that are not biodegradable accumulate as inert (or inactive) biomass (Rittmann and McCarty 2001). The amount of inert bacteria in anaerobic biomass can be quantitatively assessed with redox dyes, such as 5-cyano-2,3-ditolyl tetrazolium chloride (which targets active cells) together with fluorochrome 5-(4,6-dichlorotiazinyl) aminofluorescein (which targets all cells) (Bhupathiraju et al. 1999). Distinguishing between microbial species can be accomplished by using methods that detect and identify ribosomal RNA (rRNA) (Oude Elferink et al. 1998). Huang et al. (2002, 2003) applied rRNA methods to landfill leachate.

Aspects modelled

To simplify the complex chemistry of landfill leachate, attention is focused on the fate and transport of the three primary substrates for biomass growth: acetate, propionate, and butyrate. Methanogenesis of acetate and acetogenesis of propionate and butyrate are simulated because both processes generate biomass; the latter process also produces acetate.

The concentrations of these acids are represented by $C_A$, $C_P$, and $C_B$ (acetate, propionate, and butyrate, respectively) here.

It is assumed that each of these VFAs contributes to the growth of a separate biomass, which occurs in two parts: one that is attached to the porous medium and quantified in the model by biofilm thickness; and one that is suspended in the leachate and quantified by its suspended concentration. Film thickness and suspended concentrations are represented by $L_{\text{LAD}}$ and $C_{\text{AD}}$ (acetate degraders), $L_{\text{LDP}}$ and $C_{\text{PD}}$ (propionate degraders), and $L_{\text{LBBD}}$ and $C_{\text{BD}}$ (butyrate degraders); the total thickness of the substrate degrader film is $L_{\text{LSD}}$. Exchange of solids between suspended and attached phases is predicted through attachment (suspension to film) and detachment (film to suspension) rates. Endogenous decay of the attached and suspended biomass is included, and a fraction of the decayed active biomass is converted to inert biomass, which accumulates in a separate film (with thickness $L_{\text{LB}}$) or suspension (with concentration $C_{\text{IB}}$). The source leachate may also contain suspended biomass that is separated into active and inert components. Figure 1 illustrates the species pathways modelled (but does not show the production of acetate). This figure will aid the reader in understanding further discussions of species fate. Details about how these mechanisms are simulated are given later.

**Accumulation of CaCO3**

**Background and rationale**

The investigation of the drainage systems of 10 sanitary landfills by Brune et al. (1991) revealed cemented mineral clog matter (in addition to soft organic matter) at nearly all sites. The average composition of this material (by dry mass) was approximately 34% carbonate, 21% calcium, 16% silicon, 8% iron, 2.5% sulfur, and 1% magnesium. This clog matter, which was mainly CaCO3 and iron sulfide, significantly reduced the hydraulic transmissivity of the drainage systems examined. Likewise, the exhumation of a Canadian landfill drainage system described by Fleming et al. (1999) also showed considerable buildup of cemented mineral deposits. The composition of the Canadian clog material was similar to that found in German landfills: 30% carbonate,
20% calcium, 21% silicon, 2% iron, and 5% magnesium. Manning (2000) and Maliva et al. (2000) also reported that CaCO3 dominated LCS clog samples. This direct evidence indicates that the dominant inorganic constituent in landfill clog matter is CaCO3.

Brune et al. (1991) found that hard clog matter had formed most rapidly at sites where organic and inorganic loading was greatest. On the basis of the analysis of landfill leachate, gases, deposits, and organic matter and tests conducted in situ with exposed glass slides and gravel, Brune et al. (1991) hypothesized that the metabolic activity of microorganisms was causing the formation of the hard clog matter on the surface of the bacteria. Using data from landfill leachate experiments, Rittmann et al. (1996) provided a theoretical framework for the formation of CaCO3 in landfill drainage systems. They indicated that when the VFAs in landfill leachate (primarily acetic acid) are degraded by microbes, some of the resultant carbon dioxide dissolves to become carbonic acid. This results in an increase in pH and total carbonates, causing or accelerating the precipitation of CaCO3. The Rittmann et al. (1996) global geochemistry approach differs from that of Brune et al. (1991), in that it allows precipitation to occur away from the surface of the bacteria. Substantiating the Rittmann et al. (1996) approach, Bordier and Zimmer (1999) observed precipitates containing calcium, iron, and sulfur forming on sites not colonized by bacteria, in experiments using landfill leachate. Maliva et al. (2000) took the stance that CaCO3 crystal nucleation may be directly controlled by bacterial activity, whereas the crystal growth is largely abiotic.

Aspects modelled

To represent CaCO3 accumulation, it is necessary to model the transport of dissolved calcium and the precipitation of CaCO3. Because microbial activity drives the CaCO3 precipitation rate, the predicted rates of methanogenesis and acetogenesis will be used to derive the rate of precipitation, as described later. Since the percentage of CaCO3 in the clog matter has been found to be stable and dominant, relative to other minerals (Brune et al. 1991; Fleming et al. 1999), the model uses the rate of CaCO3 precipitation as the basis for predicting the precipitation of all locally precipitated solids. On the basis of both field and laboratory observations, the CaCO3 and the other precipitates are assumed to form a film of inorganic solids on the porous media in the drainage system. In the model, the concentration of dissolved calcium is represented by \( C_{Ca} \), and the thickness of the inorganic solids film is represented by \( L_{film} \).

Accumulation of suspended inorganic particles

Background and rationale

Clog samples reported by Fleming et al. (1999) contained a significant fraction of sand, fine gravel, and other material not usually present in the coarse gravel drainage layer. In addition, Manning and Robinson (1999) monitored leachate composition and found that the suspended solid composition included various minerals, some of which were likely from waste or daily cover, and CaCO3 was consistently present in leachate samples. Rowe et al. (2002) compared column experiments using real leachate (which naturally contained both suspended organic and suspended inorganic particles) with those using synthetic leachate (which did not contain a significant amount of these solids). Even though the synthetic leachate had a higher mass loading of organic acids, the columns with real leachate experienced a more rapid decrease in measured porosity. Rowe et al. concluded that this increase in clogging was due to the suspended solids content, likely because it readily attaches to the porous media and provides nucleation sites for further growth. Suspended inorganic particles were a significant component of clog matter in field cases, with silica representing 16%–21% of the clog material, but have been less significant (2%–4%) in the clog matter formed in laboratory column experiments with real leachate (because soil particles may settle out before testing) and negligible in synthetic leachate-produced clog matter (0%).

Aspects modelled

Because suspended inorganic particles significantly affect the rate of clogging of porous media, the model predicts the transport and attachment of these particles to the inorganic solids film, as described later. The concentration of these particles is represented by \( C_{IP} \).

Summary

As illustrated in Fig. 1, the BioClog model considers the transport and fate of propionate, acetate, butyrate, suspended propionate degraders, suspended acetate degraders, suspended butyrate degraders, dissolved calcium, suspended inorganic particles, and suspended inert biomass. The clog matter consists of five distinct components, each represented by a separate film layer: propionate degraders, acetate degraders, butyrate degraders, inert biomass, and an inorganic layer composed of mineral precipitates and other inorganic solids.

Calculation of clog thickness, porosity, and species reaction terms

Microbial growth kinetics

Certain microbial growth kinetic parameters and models are widely used in wastewater treatment and other research involving microbial growth. The most commonly used mathematical model for the dependence of the rate of microbial utilization (consumption) of substrate \( U \) on substrate concentration \( C \) is attributed to Monod (1949):

\[
U = \frac{\dot{q}C}{K_S + C}
\]

where \( \dot{q} \) is the maximum specific rate of substrate utilization; and \( K_S \) is the half-maximum rate substrate concentration. The equation was derived from an empirical analysis of microbial growth data. This equation is used for both biofilm and suspended biomass VFA utilization in BioClog. The rate of loss of microbial cell mass is quantified by the endogenous decay rate \( (b_d) \), and the relationship between the rate of microbial growth and substrate utilization is quantified by the maximum yield rate \( (Y) \), which is defined as the mass of biomass produced, divided by the mass of substrate utilized.
Microbial kinetic parameters $K_S$, $q$, $b_d$, and $Y$ are specific to the substrate, can be estimated experimentally, and may be dependent on environmental conditions, such as pH, temperature, and loading rate. For this reason, reported values have wide ranges (Pavlostathis and Giraldo-Gomez 1991). See Pavlostathis and Giraldo-Gomez (1991) and Vavilin and Lokshina (1996) for summaries of reported parameter values found for the VFAs used in this work (acetate, propionate and butyrate), along with other wastewater constituents. VanGulck (2003) used BioClog to estimate $K_S$, $q$, $b_d$, and $Y$ for acetate and butyrate by calibration to data from column experiments operated with synthetic leachate.

Effect of clogging on porosity and specific surface

A representation of the porous media is required to calculate porosity and specific surface area (surface area per unit volume) as a function of the predicted clog-film thickness. The granular medium used in drainage systems typically has a fairly uniform grading curve but may range in size from sand to coarse gravel (crushed stone). The porous medium used in experimental studies of collection system clogging is usually uniformly graded gravel or glass beads. In BioClog, the porosity and specific surface area of granular material with relatively uniform grain diameters are approximated by the sphere model presented by Taylor et al. (1990) and corrected by Cooke and Rowe (1999) to better represent the effects of the thicker films found in clogged systems.

The method uses packed spheres of equal diameter to represent the porous media in each element. Assuming stable, regular packing of equal-sized spheres, only four arrangements exist. The most open arrangement is cubic packing, with a porosity of 47.64%; and the most dense is rhombohedral packing, with a porosity of 25.95%. Since the geometry of each of the four arrangements can be defined completely, equations can be derived for computing porosity and specific surface, given the grain size of the spheres and the thickness of the uniform film coating the spheres for each arrangement (see Cooke and Rowe 1999).

Because the film thicknesses are recalculated at each time step, the porosity and specific surface of each element must also be computed for each time step. For each element, the total film thickness is used to determine two sets of porosity and specific surface values, one for each of the fully realized packing arrangements bounding the actual initial arrangement. Linear interpolation, based on the initial porosity of the element and the initial porosities of the two bounding packing arrangements, is then performed between the two sets of values to get the new porosity and specific surface for the element.

Substrate flux into the substrate degrader films

Assuming substrate is utilized within the biofilm according to Monod kinetics and molecular diffusion, Atkinson and Davies (1974) presented an approximate solution for the flux of substrate into a biofilm ($J$) as a function of biofilm thickness ($L_o$) and substrate concentration at the biofilm surface. Rittmann and McCarty (1981) utilized this relationship and incorporated mass-transport resistance from the bulk liquid to the surface of the biofilm by assuming that an effective diffusion layer of uniform thickness existed on the surface of the biofilm. The diffusion layer is given a thickness ($L_o$), and it is assumed that 1D diffusion occurs through this layer. BioClog uses the general solution (the shallow model) developed by Rittmann and McCarty (1981) to calculate $J$ by an iterative method that is detailed in Appendix A. Each of the three active films is treated independently and has separate kinetic parameters, resulting in three substrate fluxes: $J_p$, $J_A$, and $J_b$, for propionate, acetate, and butyrate, respectively.

Detachment and total bacterial loss

Calculation of total loss rate coefficient

Bacterial matter is lost from biofilm as a result of cell decay and physical detachment. The total loss rate coefficient ($b'$), which accounts for both of these processes, is the sum of the rate coefficients for each:

$$
[3] \quad b' = b_d + b_{det}
$$

where $b_{det}$ is the rate of loss due to detachment.

Numerous methods have been published for estimating detachment rate expressions (see Peyton et al. (1995) and Stewart (1993) for examples). Peyton and Characklis (1993) suggested that the large number of methods indicates a failure of any one method to model the detachment rate over a broad range of conditions. For this reason, the authors have chosen to estimate $b_{det}$ by a combination of two published methods that are based on very different detachment mechanisms: (i) a method incorporating shear stress (Rittmann 1982); and (ii) a method in which growth rate influences detachment (Peyton and Characklis 1993). A new detachment rate is calculated for each element at each time step, and hence the value varies in both space and time.

The total loss rate coefficient ($b'$) is computed as the sum of the loss rate due to decay and the loss rate due to detachment. The detachment rate is partitioned into the detachment rate due to shear stress and that due to growth rate, in accordance with a percentage due to shear stress ($f_s$), which is an input parameter $(0 \% \leq f_s \leq 100 \%)$:

$$
[4] \quad b' = b_d + \left( \frac{f_s}{100} \right) b_s + \left( 1 - \frac{f_s}{100} \right) b_G
$$

where $b_s$ and $b_G$ are the detachment rates due to shear stress and growth, respectively, as explained in the sections below.

Rate of detachment as a function of shear stress

Rittmann (1982) developed a first-order loss term that expresses detachment as a function of shear stresses imparted on the biofilm based on experimental data collected by Trulear and Characklis (1980). Rittmann (1982) found that the shear stress on the biofilm ($\sigma$) could be estimated from

$$
[5] \quad \sigma = \frac{\mu \nu_0 (1 - n)^3}{d_g^2 n A_s (7.46 \times 10^{10})}
$$

where $\mu$ is the viscosity of water; $d_g$ is the diameter of porous media grains; and $A_s$ is the specific surface of the porous media. The rate of detachment for each film ($b_{S,Det}$), assuming the presence of no other films, is
where \( L_{t,\text{Det}} \) is the thickness of the film.

Because the original shear-stress detachment method was derived for a single film, the method was modified to better suit the multiple-film system used in this model. It may be assumed that the active degrader biofilms are completely intermixed with one another and that the inert biofilm is predominantly located beneath this layer, a distribution similar to that implemented in the multiple-species biofilm model by Furumai and Rittmann (1994). The inorganic solids film may be assumed to lie predominantly beneath the four organic layers. Under this distribution, some form of protection of the inert biofilm and the inorganic solids film from the full rate of shear detachment might be expected. Conversely, it may be assumed that all the films are intermixed and thus that all are subjected to the same detachment (no protection). The detachment rate of the active degrader biofilms, or the inert biofilm, or the inorganic solids film (if chosen to be unprotected) is not modified:

[6] \[
b_{S,\text{Det}} = \begin{cases} 
8.42 \times 10^{-2} \sigma^{0.58} \\
8.42 \times 10^{-2} \left[ \frac{\sigma}{1 + 433.2(L_{t,\text{Det}} - 0.003)} \right]^{0.58}
\end{cases}
\]

If protected, the detachment rate for the inert biofilm and the inorganic solids film is modified according to the following formula:

[7] \[
b_S = b_{S,\text{Det}}
\]

where \( L_{t,\text{Prot}} \) is the total thickness of the protecting films (= \( L_{t,SD} \) for inert biofilm and \( L_{t,SD} + L_{t,IB} \) for inorganic solids film); and \( P \) controls the amount of "protection" from detachment given to the inert or inorganic films. One example for \( P \) is \( P = 0.004 \), where the detachment rate of the inert biofilm is 10% of \( b_{S,\text{Det}} \) when the thickness of the active biofilms is 100 \( \mu \)m (as used by Furumai and Rittmann 1994).

**Rate of detachment as a function of growth**

Assuming the key process controlling the detachment rate in biofilm is related to growth rate, Peyton and Characklis (1993) calculated the detachment flux rate \( (R_d) \) from a biomass material balance on a biofilm reactor, using the following:

[9] \[
R_d = k_{\text{det}} \frac{Q}{A_b} (S_i - S_i) Y L_t
\]

where \( k_{\text{det}} \) is the detachment rate coefficient; \( Q \) is the volumetric flow rate; \( A_b \) is the biofilm surface area; \( S_i \) is the substrate concentration in the influent to the reactor; and \( S_i \) is the substrate concentration within the reactor. Peyton and Characklis (1993) showed agreement between this expression and experimental data for a \( k_{\text{det}} \) value of 63 \( \text{cm}^{-1} \) for a mixed population biofilm. In this model, the expression is rewritten as

[10] \[
b_G = \frac{63 JY}{X_t}
\]

where \( b_G \) is the detachment rate due to growth rate effects; and \( X_t \) is the density of the biofilm. For the inert biofilm or inorganic solids film, which does not grow biologically, \( b_G \) is zero.

**Attachment**

**Attachment coefficient**

Suspended substrate degraders, suspended inert biomass, and suspended inorganic particles may be removed from the leachate through the process of attachment to the porous media. According to Hozalski and Bouwer (1998), bacteria reach surfaces by (i) diffusive transport (Brownian motion); (ii) advective transport (interception); (iii) sedimentation by gravity; and (iv) active cell movement (motility). Attachment of suspended solids can be expressed by a first-order rate coefficient, the attachment coefficient \( (K_{\text{Det}}) \) (Tien and Payatakes 1979; Ives 1982). In BioClog, two methods are available for calculating the coefficient: a particle filtration method, from Rajagopalan and Tien (1976); and a network method, from Reddi and Bonala (1997). The choice is left to the model user. These methods are briefly explained in the next two sections. The rate coefficients are recalculated at every time step for each element and for each species that undergoes attachment.

**Rajagopalan and Tien filtration model**

Yao et al. (1971) first published an expression for single-collector efficiency \( (\eta) \), which was further modified by Rajagopalan and Tien (1976), giving the RT model. The single-collector efficiency expresses the fraction of suspended particles moving near a collector that actually collides with the collector. The RT model accounts for the processes of diffusion, interception, sedimentation, hydrodynamic retardation, and London – van der Waals attraction. The single-collector efficiency is computed with

\eta = 1.5 H_p (1 - n)^{2/3} N_{R}^{2} \left[ \frac{N_{LO}^{1/8} N_{R}^{-1/8} + (2.25 \times 10^{-3})}{N_{G}^{1/2} N_{R}^{-2/4} + 4 (1 - n)^{2/3} H_p^{1/3} N_{Pe}^{-2/3}} \right]
\]

where \( H_p \) is the Happel parameter; \( N_{R} \) is the interception parameter; \( N_{LO} \) is the London force parameter; \( N_{G} \) is the gravitational parameter; and \( N_{Pe} \) is the Peclet number. Details on the calculation of these parameters are given by Tien (1989). Numerous researchers have used the RT model to predict and study the removal of suspended particles by granular fil-

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ers (see Tobiason and O’Melia 1988; Taylor and Jaffè 1990; Hornberger et al. 1992; Martin et al. 1992; Logan et al. 1995; Putnam and Burns 1997). It should be noted that in the above computations, the model allows the particle diameter and density of suspended solids to differ between those that are active degraders, those that are inert biomass, and those that are inorganic, which allows the use of unique values of $\eta$ for each type of suspended particle.

The attachment coefficient ($K_{\text{At}}$) can be computed from the collector efficiency by using the following expression (Clement et al. 1997; Hornberger et al. 1992; Tien et al. 1979):

$$K_{\text{At}} = \frac{3(1-n)V_3}{2d_g}$$  \[12\]

**Reddi and Bonala network method**

To obtain an expression for the particle deposition rate coefficient, Reddi and Bonala (1997) drew on the network simulations of Rege and Fogler (1988), who based their approach on the particle-capture probability approach proposed by Stein (1940). In this method, the probability of particle capture depends on the pore radius size ($r_p$) and a lumped parameter ($\theta_{\text{RB}}$) that accounts for gravitational, inertial, hydrodynamic, electric double layer, and van der Waals forces. The conditions for deposition become more favourable with increasing $\theta_{\text{RB}}$, which can be estimated based on the research by Rege (1988) and Rege and Fogler (1988). Calculation of the attachment rate coefficient requires the prior calculation of the pore radius ($r_p$) and the effective length of the pore tubes ($\alpha_{\text{RB}}$).

**Estimation of pore size radius**

The equation for hydraulic radius ($r_h$), translated for sphere assemblages, is (Graton and Fraser 1935)

$$r_h = \frac{V_s}{A_{\text{Surf}}} \frac{n}{1-n}$$  \[13\]

where $V_s$ and $A_{\text{Surf}}$ are the volume and the surface area of the spheres. For pipes with circular cross sections, the hydraulic radius is equal to half the actual radius of the pipe (Graton and Fraser 1935). For this attachment method, pores are considered to act as pipes with constant circular cross sections; thus, the pore radius ($r_p$) is

$$r_p = 2r_h$$  \[14\]

Combining eqs. [13] and [14] results in

$$r_p = \frac{d_g}{3} \frac{n}{1-n}$$  \[15\]

**Estimation of effective pore tube length**

The effective pore tube length, $\alpha_{\text{RB}}$, is a length parameter associated with each particle (Reddi and Bonala 1997). It is estimated by rearrangement of an expression by Arya and Dierolf (1989), who assumed uniformly sized spheres in a cubic packing arrangement:

$$\alpha_{\text{RB}} = \frac{n}{1-n} \frac{d_g^3}{6r_p^3}$$  \[16\]

**Calculation of attachment coefficient**

The attachment rate coefficient is calculated with the following (Reddi and Bonala 1997):

$$K_{\text{At}} = \frac{V_{pw}}{4\alpha_{\text{RB}}} \left[ 4 \left( \frac{k_{\text{RB}}}{r_p} \right)^2 - 4 \left( \frac{k_{\text{RB}}}{r_p} \right)^4 + \left( \frac{k_{\text{RB}}}{r_p} \right)^4 \right]$$  \[17\]

where $V_{pw}$ is the pore-water velocity; and $k_{\text{RB}} = 0.5\eta d_p$, where $d_p$ is the particle diameter of the suspended solids. It should be noted that the particle size and lumped parameter might differ between suspended solids that are active, inert, and inorganic. In the present context, $r_p$ and $\alpha_{\text{RB}}$ (eqs. [15] and [16]) are calculated on the basis of the current (clogged) porosity.

**Accumulation of clog matter as films**

**Thickness of substrate degrader film**

The film thickness of each of the three substrate degraders is computed for each element. Given the rate of substrate flux into the biofilm ($J$) and the rate of total loss ($b'$, computed from eq. [3]) for each specific substrate, the change in film thickness of the substrate degrader due to growth and loss of biofilm during one time step is computed according to a method developed by Rittmann and Brunner (1984). Using propionate as an example, one can use the following to compute the change in film thickness due to growth:

$$L_{f,PD,\text{Growth}} = \frac{J_p X_p}{b_{\text{PD,SD}}} \Delta t$$  \[18\]

where $Y_p$ is the maximum yield coefficient for propionate. The decrease in thickness due to biomass loss is

$$L_{f,PD,\text{Loss}} = b'_{pd} L_{f,PD,\text{Prev}} \Delta t$$  \[19\]

where $b'_{pd}$ is the total loss rate coefficient for propionate degraders; and $L_{f,PD,\text{Prev}}$ is the film thickness of the propionate degrader film for this element in the previous time step. The change in film thickness due to attachment of suspended degraders is given by

$$L_{f,PD,\text{Attach}} = \frac{nK_{\text{At},PD} C_{PD,Avg}}{A_s X_{f,SD}} \Delta t$$  \[20\]

where $C_{PD,Avg}$ is the average suspended propionate degrader concentration for the element (from the concentration at each node).

The density of the substrate degrader biofilms and inert biofilm can be selected to be either dissimilar and constant or a function of porosity decrease, according to the following relationship:

$$X_{f,SD} = X_{f,IB} = A_X \frac{n_i - n}{n_i} + B_X$$  \[21\]

where $n_i$ is the initial, clean media porosity; and coefficients $A_X$ and $B_X$ are calculated from linear regression using experimental data, where biomass density and porosity have been measured. VanGulck (2003) estimated coefficients of 247 and 72 mg VS cm$^{-3}$ (where VS is volatile solids, a measure
of organic solids) for $A_x$ and $B_x$, respectively, for columns operated with real leachate.

The final film thickness for propionate degraders after each time step is computed by using

\[ L_{f,PD} = L_{f,PD,Prev} \left( \frac{X_{f,SD,Prev}}{X_{f,SD}} \right) + L_{f,PD,Growth} - L_{f,PD,Loss} + L_{f,PD,Attach} \]

where $X_{f,SD,Prev}$ is the substrate degrader film density of the previous time step. The ratio of previous film density to current film density allows the film thickness to be modified to account for densification of the film if the option to have variable density is applied. Calculations similar to eqs. [18]–[20] and [22] are performed to update the thicknesses of the acetate and butyrate films.

**Thickness of inert biofilm**

Although the inert biofilm does not grow (in the biological sense), it gains matter through attachment of the suspended inert biomass and from the decay of the active films, and it loses matter through detachment. At each time step, some fraction of the active biomass undergoes decay but is not lost from the system. The fraction that is not biodegraded is $1 - f_b$, where $f_b$ represents the fraction that is degradable for all substrate degraders. Biodegradation research discussed by Rittman and McCarty (2001), for example, showed that approximately 20% of biomass was never biodegraded; thus, for these experiments, $f_b = 0.8$. The fraction that decays and is not biodegraded is added to the inert biofilm. The change in thickness representing this transformation from active to inert is

\[ L_{f,IB,Inert} = (b_{d,AD} L_{f,AD,Prev} + b_{d,PD} L_{f,PD,Prev} + b_{d,BD} L_{f,BD,Prev}) (1 - f_b) \frac{X_{f,SD,Prev}}{X_{f,IB}} \Delta t \]

where $b_{d,AD}$, $b_{d,PD}$, and $b_{d,BD}$ are the decay coefficients for acetate, propionate, and butyrate, respectively. As with the active films, the change in thickness due to biomass loss is

\[ L_{f,IB,Loss} = b_{ib} L_{f,IB,Prev} \Delta t \]

where $b_{ib}$ is the total loss rate coefficient for the inert biofilm; and $L_{f,IB,Prev}$ is the previous thickness of the inert biofilm. The change in film thickness due to attachment is

\[ L_{f,IB,Attach} = \frac{n K_{AI,IB} C_{IB,Avg}}{A_X X_{f,IB}} \Delta t \]

where $K_{AI,IB}$ is the attachment coefficient for the suspended inert biomass; and $C_{IB,Avg}$ is the average suspended inert biomass concentration for the element.

The inert biofilm thickness is then calculated by using

\[ L_{f,IB} = L_{f,IB,Prev} \left( \frac{X_{f,IB,Prev}}{X_{f,IB}} \right) + L_{f,IB,Inert} - L_{f,IB,Loss} + L_{f,IB,Attach} \]

**Thickness of inorganic solids film**

The inorganic solids film gains matter from the accumulation of CaCO$_3$, other locally precipitated solids (with an assumed precipitation rate proportional to that of CaCO$_3$), and attachment of suspended inorganic particles; and it loses matter to detachment. The flux of CaCO$_3$ onto the porous media, $J_c$, is computed by using

\[ J_c = -2.5 F_{R,Ca,Avg} \frac{n}{A_X L_c} \]

where $F_{R,Ca,Avg}$ is the average calcium flux for the element (calculated from eq. [42]); the multiplier 2.5 converts the calcium flux to CaCO$_3$ flux; and $L_c$ is the length of the element. From the flux of CaCO$_3$ ($J_c$), the implicit change in film thickness due to CaCO$_3$ accumulation is

\[ L_{f,IS,CaCO3} = \frac{J_c}{X_{f,IS}} \Delta t \]

The model allows for the accumulation of matter that is not CaCO$_3$ at a rate directly related to the rate of CaCO$_3$ accumulation. The change in thickness due to this accumulation is given by

\[ L_{f,IS,OP} = f_{OP} L_{f,IS,CaCO3} \]

where $f_{OP}$ may be defined in two ways. (i) If the attachment of inorganic solids is being modelled by one of the given methods, then $f_{OP}$ is the ratio of the volume of inorganic solids (not CaCO$_3$) to the volume of CaCO$_3$ that are precipitated locally (the subscript OP means other precipitates). Because this fraction excludes inorganics from suspension, the value of $f_{OP}$ should be calculated from analysis of clog matter grown from leachate that is very low in suspended solids content, such as synthetic leachate. In VanGulick and Rowe (2004b), for example, an average value of 0.06 for $f_{OP}$ can be calculated from the measured clog properties, which illustrates the dominance of CaCO$_3$ precipitation over other precipitation. (ii) If the attachment of inorganic solids is not being modelled by one of the given methods, then $f_{OP}$ can be used to estimate the net accumulation of inorganic solids that are not CaCO$_3$ at each time step, assuming they accumulate through precipitation and attachment, at a rate proportional to the precipitation of CaCO$_3$. In this case, $f_{OP}$ would be defined as the ratio of the volume of the inorganic solids (not CaCO$_3$) to the volume of CaCO$_3$, without regard to the source of the non-CaCO$_3$ inorganic solids. This value would be expected to be larger, as it accounts for all attachment of inorganic solids, along with precipitation of other precipitates.

As with the other films, the change in thickness due to the loss of inorganic solids is

\[ L_{f,IS,Loss} = b_{IS} L_{f,IS,Prev} \Delta t \]

where $b_{IS}$ is the total loss rate coefficient for the inorganic solids film; and $L_{f,IS,Prev}$ is the previous thickness of the inorganic solids film.

The attachment of suspended inorganic particles is controlled by the rate of attachment of suspended inorganic particles ($K_{Att,IP}$) and the concentration of suspended inorganic particles in the element, which is the average of the concen-
trations at the bounding nodes, \( C_{IP,AVG} \). The increase in thickness due to attachment of suspended inorganic solids is

\[
L_{f,IS,Attach} = \frac{nK_{AIl,IP}C_{IP,AVG}}{A_sX_{f,IS}} \Delta t
\]

The new thickness of the inorganic solids film is calculated by using

\[
L_{f,IS} = L_{f,IS,Prev} + L_{f,IS,CO_3} + L_{f,IS,OP} - L_{f,IS,Loss} + L_{f,IS,Attach}
\]

where \( L_{f,IS,Prev} \) is the thickness of the inorganic solids film computed in the previous time step.

**Species reaction terms**

**Reaction contribution and flux terms**

The variable \( S_R \) is the rate of addition or removal of each species calculated on the basis of the physical and biogeochemical reactions modelled. The contribution of these reactions to the total reaction flux, for each species \( i \) at each node, is

\[
R_i = \beta S_R
\]

where

\[
\beta = \frac{nL_x}{2}
\]

(two-noded linear element)

Because the reaction terms require both nodal properties (e.g., species concentration) and elemental properties (e.g., porosity), a separate \( R_i \) value is calculated for each connected node–element combination. This results in two contributions to the total reaction flux at the node (a contribution from the elements on each side of the node). The sum of these contributions for each node constitutes the reaction portion of the point sink (loss) or source (gain) flux terms used by the transport model. The derivations of the reaction flux contribution terms for each species \( (R_i) \) are explained next.

**Substrate (volatile fatty acid) reactions**

A non-steady-state biofilm growth model (Rittmann and McCarty 1981) is used to estimate the acetogenesis of propionate and butyrate and the methanogenesis of acetate by the substrate degrader films, resulting in estimates of the substrate flux into the biofilm, \( J_p, J_A \), and \( J_B \). Substrate degrada
tion by the suspended degraders is predicted on the basis of Monod kinetics. The equations for the contributing fluxes for the three substrates are as follows:

\[
R_p = \beta \times \left( \frac{J_p A_s}{n} - \frac{\dot{q}_p C_{PD} C_p}{K_{S,p} + C_p} \right)
\]

\[
R_A = \beta \times \left( \frac{J_A A_s}{n} + \frac{\dot{q}_A C_{AD} C_A}{K_{S,A} + C_A} + 0.5714 \left( \frac{J_B A_s}{n} - \frac{\dot{q}_B C_{BD} C_B}{K_{S,B} + C_B} \right) \right)
\]

\[
R_B = \beta \times \left( - \frac{J_B A_s}{n} - \frac{\dot{q}_B C_{BD} C_B}{K_{S,B} + C_B} \right)
\]

where \( \dot{q}_p, \dot{q}_A, \) and \( \dot{q}_B \) are the maximum specific rates of substrate utilization for propionate, acetate, and butyrate; and \( K_{S,p}, K_{S,A}, \) and \( K_{S,B} \) are the half-maximum rate substrate concentrations for propionate, acetate, and butyrate. The values 0.5714 and 0.8 are the ratios of the mass of acetate created to the mass of propionate and butyrate degraded, respectively.

The terms of eqs. [35]–[37] can be briefly explained by taking eq. [36] as an example. In this equation, the first two terms after the first bracket represent the removal of acetate due to utilization by the acetate degrader film and by the suspended acetate degraders, respectively. The third and fourth terms represent the production of acetate due to propionate utilization by the propionate degrader film and by the suspended propionate degraders, respectively. The fifth and sixth terms represent the production of acetate due to the utilization of butyrate by the butyrate degrader film and by the suspended butyrate degraders.

**Suspended substrate degrader reactions**

The net rate terms for the three types of suspended biomass are

\[
R_{PD} = \beta \times \left( \left( b'_{PD} - b_{d,PD} \right) \frac{X_{f,SD} L_{f,PD} A_s}{n} - K_{AIl,PD} C_{PD} + \frac{Y_p \dot{q}_p C_{PD} C_p}{K_{S,p} + C_p} - b_{d,PD} C_{PD} \right)
\]

\[
R_{AD} = \beta \times \left( \left( b'_{AD} - b_{d,AD} \right) \frac{X_{f,SD} L_{f,AD} A_s}{n} - K_{AIl,AD} C_{AD} + \frac{Y_A \dot{q}_A C_{AD} C_A}{K_{S,A} + C_A} - b_{d,AD} C_{AD} \right)
\]

\[
R_{BD} = \beta \times \left( \left( b'_{BD} - b_{d,BD} \right) \frac{X_{f,SD} L_{f,BD} A_s}{n} - K_{AIl,BD} C_{BD} + \frac{Y_B \dot{q}_B C_{BD} C_B}{K_{S,B} + C_B} - b_{d,BD} C_{BD} \right)
\]

where \( b'_{PD}, b'_{AD}, \) and \( b'_{BD} \) are the total loss coefficients for propionate, acetate, and butyrate attached films for the element; \( K_{AIl,PD}, K_{AIl,AD}, \) and \( K_{AIl,BD} \) are the attachment coefficients for the suspended propionate degraders, suspended acetate degraders, and suspended butyrate degraders; and \( Y_p, Y_A, \) and \( Y_B \) are the maximum yield coefficients for propionate, acetate, and butyrate.

**Suspended inert biomass reactions**

The net rate term of the suspended inert biomass is
Precipitation of CaCO$_3$

Calcium is only removed from the leachate by precipitation as CaCO$_3$. Determination of the rate of CaCO$_3$ precipitation is based on the premise that the mass of calcium precipitated out of leachate as CaCO$_3$ is directly related to the net production of carbonic acid in the system (see VanGulck et al. 2003). This is a refinement of the previous technique adopted by Cooke et al. (1999, 2001) that related CaCO$_3$ precipitation to removal of chemical oxygen demand (COD).

The rate term for calcium removal at each node of each connected element is

\[ R_{\text{Ca}} = \beta \times ( - Y_H R_{\text{H,CO}_3} ) \]

where \( Y_H \) is the carbonic acid (H$_2$CO$_3$) yield coefficient; and \( R_{\text{H,CO}_3} \) is the rate of production of carbonic acid:

\[ R_{\text{H,CO}_3} = 0.9684 \left( \frac{J_A A_s}{n} + \frac{\dot{q}_A C_{AD} C_A}{K_{S,A} + C_A} \right) + 0.1384 \left( \frac{J_P A_s}{n} + \frac{\dot{q}_P C_{PD} C_P}{K_{S,P} + C_P} \right) - 0.19375 \left( \frac{J_B A_s}{n} + \frac{\dot{q}_B C_{BD} C_B}{K_{S,B} + C_B} \right) \]

and \( Y_H \) is an empirically derived coefficient, defined as

\[ Y_H = \frac{\text{Ca}^{2+} \text{ removed}}{\text{net H}_2\text{CO}_3 \text{ produced}} \]

The values 0.9684 and 0.1384 are the mass ratios of carbonic acid produced to acetate and propionate fermented, respectively; 0.19375 is the mass ratio of carbonic acid consumed to butyrate fermented, with the quantity fermented in units of mg COD L$^{-1}$ (VanGulck et al. 2003). An estimate of \( Y_H \) for use in modelling can be made either by adopting a published value or by determining a value experimentally. In either case, the leachate composition, along with the conditions for biomass growth and mineral precipitation, must approximate the system being modelled. Experimental estimation of \( Y_H \) requires the measurement of calcium and VFA concentrations at positions before and after (and within, if possible) the zone of biomass growth, as well as the measurement of CaCO$_3$ precipitation at numerous times over the period of operation. The experiment should not be terminated until the rapid growth phase has ended (normally at a period of near steady state). The coefficient \( Y_H \) can then be estimated by plotting the calcium concentration removed versus net H$_2$CO$_3$ produced at each measurement time and then performing linear regression through the data (see VanGulck et al. 2003). The net H$_2$CO$_3$ produced can be computed by using the following (VanGulck et al. 2003):

\[ \text{net H}_2\text{CO}_3 \text{ produced} = 0.9684( \text{acetate fermented}) + 0.1384( \text{propionate fermented}) - 0.19375( \text{butyrate fermented}) \]

where the values 0.9684, 0.1384, and 0.19375 are as defined for eq. [43]. It can be seen from these ratios that the fermentation of acetate is driving the production of carbonic acid and thus the rate of precipitation of CaCO$_3$.

Because the calcium sink term depends only on the predicted microbial activity (carbonic acid production) of the system and not on the amount of calcium available, values may be computed that, within the transport calculation, remove more calcium than exists at the node. Unchecked, this excessive value would cause both a negative calcium concentration and a precipitation rate that is higher than physically possible for that node. For these reasons, for situations where the concentration would otherwise go below zero, the sink term is recalculated so that the removal of calcium is limited to that which causes the concentration to be zero.

Reactions of suspended inorganic particles

The net rate term for suspended inorganic particles is

\[ R_{\text{IP}} = \beta \times \left( b_{\text{IS}} \frac{X_{\text{L,IS}} A_s}{n} - K_{\text{A,IP}} C_{\text{IP}} \right) \]

Feature implementation

As the solution is marched forward in time, new clog accumulation and rates of species conversion are computed and used to modify the porosity (and specific surface) and species transport source or sink terms, respectively, at each time step. The computations are performed in the following order:

1. Substrate flux into substrate degrader films: The biofilm model is used to compute the rate of substrate utilization by each of the three substrate degrader films.
2. Detachment and total bacterial loss rate: The detachment rates are calculated for the films.
3. Attachment: The rates of attachment of the suspended solids to their respective films are calculated.
4. Film accumulation: On the basis of the rates above and other factors, the change in thickness of the inorganic and four organic films is computed.
5. Porosity and specific surface: The new film thicknesses are used to calculate the porosity and specific surface, using a geometric model.
6. Species net rate terms: The reaction rates are used in the computation of the solute transport source or sink (species flux) terms.
7. Transport: A transient finite-element model is used to predict the transport of each of the species.

Model demonstration

For the purposes of this paper, two hypothetical column experiments in which porous media is permeated with real leachate are used to illustrate some of the features of the model. Both examples model a 5.08 cm diameter column
filled with 0.6 cm glass beads permeated with real leachate at a flow rate of 1 L day⁻¹. The examples are the same, except that in case 2 the influent concentrations of suspended substrate degraders and inorganic solids are 50% of those for case 1. The model runs assume constant influent species concentrations and flow rates.

The model inputs are listed in Tables 1–3. The derivation of these parameters can be found in VanGulck (2003), except for the influent species concentrations, flow rate, and propionate kinetic constants, which were realistic values for these types of experiments (Cooke et al. 1999; VanGulck 2003). In these examples, BioClog was used to calculate results for 200 days of operation. The method used by Rajagopalan and Tien (1976) was chosen to model attachment.

Figures 2a and 2b show the influent and effluent concentrations of the nine species for case 1. Figure 2a shows decreasing effluent concentrations for the three VFAs (propionate, acetate, and butyrate) because of biological utilization; decreasing calcium as a result of CaCO₃ precipitation; and decreasing suspended inorganic solids because of their attachment. In Fig. 2b, the influent concentration of each of the substrate degraders is 58.3 mg VS L⁻¹ (assuming the influent concentration of substrate degraders (175 mg VS L⁻¹) is equally distributed between the three VFA-degrading species). The net effect of attachment, detachment, and growth of suspended substrate degraders within the column is to decrease the effluent concentration of the suspended microorganisms, even though each type of microorganism is growing as biofilm. The inert biomass only undergoes attachment and detachment.

Figure 3a shows the predicted cumulative film thicknesses for the five films at 200 days for case 1. Note that the substrate degrader biofilms are intermixed (but quantified by individual thicknesses, as shown here). It can be seen that the films are thicker at the influent end of the column (X = 0 cm) than at the effluent end (X = 30 cm). Butyrate (average thickness, 0.0040 cm) can be seen to be as significant as acetate (average thickness, 0.0046 cm) in contributing to biomass growth whereas the growth of propionate degraders (average thickness, 0.00054 cm) is slow. The inorganic solids film and the inert biofilm are the most significant portions of the clog material as a whole, having average thicknesses of 0.012 and 0.013 cm, respectively. This plot

<table>
<thead>
<tr>
<th>Table 1. Model parameters for hypothetical example.</th>
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<tr>
<td><strong>System parameters</strong></td>
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<td>Length of column</td>
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<tr>
<td>Diameter of column</td>
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<tr>
<td>Porous media diameter</td>
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<tr>
<td>Initial porosity, ( n_i )</td>
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<tr>
<td>Flow rate</td>
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<td>Longitudinal dispersivity, ( \alpha_L )</td>
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<td><strong>Problem discretization</strong></td>
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<tr>
<td>Time step length, ( \Delta t )</td>
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<td>Element length, ( L_e )</td>
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<td><strong>Leachate characteristics</strong></td>
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<td>Propionate concentration</td>
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<tr>
<td>Acetate concentration</td>
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<tr>
<td>Butyrate concentration</td>
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<tr>
<td>Ca²⁺ concentration</td>
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<tr>
<td><strong>Clog matter parameters</strong></td>
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<td>( Y_H ) of propionate</td>
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<td>( X_{LS,SD} ) and ( X_{LB,IB} )</td>
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<td><strong>Suspended solids parameters</strong></td>
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<td>Active and inert diameter</td>
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<td>Active and inert density</td>
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<td>Inorganic particle diameter</td>
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<td>Inorganic particle density</td>
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**Note:** Film densities \( X_{LS,SD} \) and \( X_{LB,IB} \) are mass of volatile solids (VS) per volume of organic film; \( X_{LS} \) is mass of nonvolatile solids (NVS) per volume of inorganic solids; and suspended solids densities are mass per unit volume. COD, chemical oxygen demand.
captures the distribution of clog matter constituents after significant mineral precipitation and inert biomass accumulation have occurred. At earlier times (not shown), the predicted inorganic solids and inert biofilm thicknesses are much less significant relative to the thicknesses of the active degraders. Figure 3b shows that the porosity is lowest near the inlet and decreases with time, particularly near the inlet.

Figure 4a shows the predicted cumulative film thicknesses for case 2, in which the influent concentrations of substrate degraders and inorganic suspended solids were reduced by 50% from those of case 1. Although the trends are similar, the thicknesses of the films are roughly half of those in case 1. Consequently, the predicted porosities are greater in case 2 than in case 1, as shown in Fig. 4b.

The reductions in film thicknesses in case 2 illustrate the significance of attachment of suspended solids in the formation of the films that constitute clog matter. First, the lower concentration of suspended substrate degraders available for attachment is directly decreasing the substrate degrader biofilm thicknesses. This decrease in active biofilm thick-
ness further decelerates growth by decreasing VFA utilization (up to the point that detachment becomes significant). Second, inert biofilm accumulation is also directly decreased by the decrease in the suspended inert biomass available for attachment. In addition, because there is less substrate degrader biofilm, the inert biofilm gains less from the decay of active substrate degraders. Lastly, since the precipitation of CaCO₃ is largely driven by the utilization of acetate, and the rate of utilization is lower in case 2, the rate of precipitation is decreased. Furthermore, the lower concentration of suspended inorganic particles available for attachment also drives down the accumulation of inorganic solids film. All these effects result in porosities not being reduced to the same extent as in case 1, where suspended solid content was high.

These examples demonstrate some of the significant features that have been incorporated into the model. In particular, the modelling of the attachment of suspended substrate...
degraders, suspended inert biomass, and suspended inorganic particles can have a significant impact on the predicted film thicknesses and porosities and will be critical for modelling of real leachate. The modelling of butyrate as a substrate and the separate inert biofilm layer are also important features of the model described here.

Conclusions

A model has been presented to predict landfill leachate transport and clogging of porous media, primarily for comparison with laboratory experiments simulating LCSs. The model allows for flow and transport, using the finite-element method. It considers the components of real leachate required to model clogging: acetate, propionate, butyrate, suspended acetate degraders, suspended acetate, propionate, and butyrate degraders, suspended inert biomass, dissolved calcium, and suspended inorganic particles. The model differentiates between active and inert biomass, and it divides the clog film into five attached films: a propionate-degrading film, an acetate-degrading film, a butyrate-degrading film, an inert biofilm, and an inorganic solids film. A biofilm model controls growth of the substrate-degrading films. A combination of two published methods is used to predict the detachment rate of the films. Two published attachment methods are used for prediction of attachment of suspended solids. Calcium carbonate precipitation is governed by carbonic acid production and calcium availability. The change in porosity and specific surface of the clogged media are predicted with the use of a geometric sphere model based on the calculated film thicknesses. Finally, an example demonstrates some of the significance of the model’s new features.

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References


Appendix A. Calculation of substrate flux, J

Atkinson and Davies (1974) presented an approximate solution for flux of substrate into a biofilm (J) as a function of biofilm thickness (L), and substrate concentration at the biofilm surface (C_f), assuming Monod kinetics and molecular diffusion control substrate utilization within the biofilm:

\[ J = \eta \bar{q} X_f L_f \frac{C_f}{K_S + C_f} \]

where \( \eta \) is the effectiveness factor; \( \bar{q} \) is the maximum specific rate of substrate utilization; \( X_f \) is the density of the biofilm; and \( K_S \) is the half-maximum rate concentration. The effectiveness factor expresses the relative significance of molecular diffusion within the biofilm as a ratio of the actual substrate flux to the substrate flux that would occur if substrate at a concentration of \( C_S \) penetrated the entire thickness of the biofilm.

To account for mass-transport resistance, Rittmann and McCarty (1981) incorporated an effective diffusion layer (a layer of stagnant liquid between the bulk fluid and the film surface) of uniform thickness (L) and assumed 1D diffusion occurs through this layer. Consequently, applying Fick’s law,

\[ J = -D_o \frac{dC}{dz} = D_o \frac{C - C_S}{L_1} \]

where \( J \) is the substrate flux per unit surface area of the biofilm; \( D_o \) is the molecular diffusivity of the substrate in the liquid; and \( C \) is the substrate concentration in the bulk fluid. Because it is assumed that some loss of substrate occurs within the liquid diffusion layer, both \( C_S \) and \( \eta \) are unknown, and thus an iterative method is required to solve eq. [A1]. The authors have adapted the general solution (the shallow model) outlined in Rittmann and McCarty (1981), which follows:

1. The transformations into dimensionless terms are

\[ C^* = \frac{C}{K_S} \]

\[ L_f^* = \frac{L_f}{\tau} \]

\[ D_f^* = \frac{D_f}{D_o} \]

\[ L^* = \frac{L_1}{\tau} \]

where \( D_f \) is the molecular diffusivity of the substrate in the biofilm; and

\[ \tau = \sqrt{\frac{2K_SD_f}{\bar{q}X_f}} \]

2. To begin the iterative method, an initial estimate of the effectiveness factor (\( \eta \)) is made using

\[ \eta = \tan h \left( \sqrt{2L_f^*} \right) \]

3. An initial estimate of the dimensionless substrate concentration at the biofilm surface (\( C_S^* \)) is calculated using the quadratic solution of the dimensionless forms of eqs. [A1] and [A2] combined:

\[ C_S^* = 0.5 \times \left[ \left( C_f - 1 - 2L_f^* D_f^* L^* \eta \right) \right. \]

\[ + \left. \sqrt{\left( C_f - 1 - 2L_f^* D_f^* L^* \eta \right)^2 + 4C_f^*} \right] \]

4. To begin the iterative loop the trial value (\( C_{S,2}^* \)) is computed using

\[ C_{S,2}^* = C_f - 2D_f^* L_f^* \eta \frac{C_f^*}{1 + C_f^*} \]

5. Also, a trial effectiveness factor (\( \eta_2 \)) is computed by using

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6. The effectiveness factors ($\eta$ and $\eta_2$) are compared for convergence. If they are not close enough, one of two methods is used to estimate $\eta$ and $C_S^*$ before returning to step 3: (i) if it is the first iteration, set $\eta = \eta_2$ and set $C_S^* = C_{S,2}^*$; or (ii) if it is not the first iteration, set $\eta = 0.5(\eta + \eta_2)$ and set $C_S^* = 0.5(C_S^* + C_{S,2}^*)$. Method (ii) was added by the authors to increase the speed of convergence.

7. Once the iterative method has converged on values of $C_S^*$ and $\eta$, the substrate flux ($J$) is found by using eq. [A1], where

$$[A10] \quad C_S = C_S^* K_S$$

References
