Macroscopic Models for Predicting Changes in Saturated Porous Media Properties Caused by Microbial Growth

by T. P. Clement, B. S. Hooker, and R. S. Skee

Abstract

Analytical equations are developed to model changes in porosity, specific surface area, and permeability caused by biomass accumulation in porous media. The proposed equations do not assume any specific pattern for microbial growth but instead are based on macroscopic estimates of average biomass concentrations. For porous media with a pore-size distribution index value ($\lambda$) equal to 3, the macroscopic model predictions of porosity, specific surface area, and permeability changes are in exact agreement with biofilm-model predictions. At other values of $\lambda$ between 2 and 5, simulated porosity profiles are identical and relative specific surface area and permeability profiles show minor deviations. In comparison to biofilm-based models, the macroscopic models are relatively simple to implement and are computationally more efficient. Simulations of biologically reactive flow in a one-dimensional column show that the macroscopic and biofilm approach based transport codes predict almost identical porosity and permeability profiles. The macroscopic models are simple and useful tools for estimating changes in various porous media properties during bioremediation of contaminated aquifers.

Introduction

In situ bioremediation is an effective aquifer restoration method that has received considerable attention in recent years. Bioremediation processes generally involve injection of appropriate nutrients to stimulate indigenous subsurface organisms. In some instances, nonindigenous microbes may be introduced to degrade specific contaminants. In both cases, as treatment progresses, accumulation of biomass in subsurface pores reduces aquifer porosity and permeability. The literature reports three distinct approaches to model microbial growth and accumulation processes in porous media. These include the continuous biofilm, discrete microcolony, and macroscopic approaches (Baveye and Valocchi, 1989).

The biofilm approach assumes continuous and uniform biomass growth on the exposed surface of each particle in a porous medium. Taylor and Jaffé (1990a, b, c) and Taylor et al. (1990) used this assumption to derive analytical expressions to model changes in porous media properties due to biological growth. However, as pointed out by Baveye and Valocchi (1989), Taylor and Jaffé’s (1990a) experimental study presented no supporting evidence to verify the actual growth patterns and film thicknesses of the attached biomass. Instead, apparent biofilm thicknesses were indirectly estimated from the permeability reductions observed in their column experiments. Cunningham et al. (1991) also proposed expressions to model the influence of biofilm accumulation on hydrodynamics in porous media. However, use of these expressions is limited to cubically packed spherical media with particles of uniform diameter.

Alternatively, Vandevivere and Baveye (1992a, b) support the microcolony model for biomass growth since they observed discrete microbial colonies in a biologically active saturated sand column. Microbial growth inside the column was sparse and heterogeneous, contrary to the basic tenets of the biofilm approach. In spite of these sparse growth patterns, considerable permeability reduction occurred in their column since accumulating bacterial aggregates formed local plugs in the pore spaces.

Both biofilm and microcolony approaches discussed above are based on a specific (continuous or patchy) type of micro-scale model for bacterial growth. In reality, growth patterns in porous media are more likely a combination of the two models, where microbes initially grow in discrete colonies and gradually expand to form continuous biofilms (Rittmann, 1993). Therefore, the macroscopic approach, the third alternative which considers only spatially averaged biomass concentrations, is a more realistic option for describing biomass in porous media. This option offers more adaptability because it does not suppose any specific growth pattern. It is also the approach most commonly reported in the literature (Corapcioglu and Haridas, 1984; Borden and Bedient, 1986; and Kindred and Celia, 1989). Moreover, in most microbiologically mediated transport studies involving porous media, experimental data for microbes are reported only as average biomass concentrations (Taylor and Jaffé, 1990a; Lozada et al., 1994; Jennings, 1994; and Lundman, 1992). These data can directly conform only to the macroscopic approach because biofilm parameters are generally estimated through a modeling framework rather than direct observations. Actual measurement of biofilm thicknesses or microcolony densities is a tedious and costly process that is often overwhelmed by experimental error. Also, these pore-scale measurements may not be useful for describing the overall fluid transport in porous media because Darcy-scale flow equations and soil property expressions used for modeling the transport are continuum models. These models are developed based on representative elementary volume averaging procedures that disregard microscopic flow in individual pores (Bear, 1979). Hence, simultaneous use of a pore-scale
(microcolony or biofilm) biological model and a Darcy-scale transport model to describe reactive transport in porous media has a conceptual disparity in problem scale.

The aim of this paper is to develop a set of analytical expressions to model changes in porosity, specific surface area, and permeability caused by biomass accumulation in porous media. We adopt the macroscopic approach and use attached biomass concentration as the model variable to describe variations in porous media properties. The proposed models do not assume a specific pattern for biomass distribution and hence they are applicable under a variety of growth conditions. A detailed comparison between the macroscopic and biofilm approaches is also presented.

Modeling Porous Media Properties

Using the cut-and-random-rejoin model as the basis for derivation, analytical expressions for permeability and specific surface area can be derived from the following mathematical relations (Taylor et al., 1990; Mualem, 1976):

\[
k = \tau G n_o \sqrt{\int_{r_o}^{R} \frac{f(r)}{r} \, dr} \]

\[
M = 2 \int_{r_o}^{R} \frac{f(r)}{r} \, dr \]

where \( k \) is the soil permeability (L²), \( M \) is the specific surface area (L⁻¹), \( G \) is a geometric factor taken to be 1/8 in Taylor et al. (1990), \( r \) is the tortuosity factor, \( n_o \) is the soil porosity, \( r \) is a pore radius, \( r_o \) is the minimum pore radius, \( R \) is the maximum pore radius, and \( f(r) \) is the Pore-Size Distribution (PSD) function that describes the distribution of pore radii. The functional form of \( f(r) \) can be derived from soil-water (drainage) retention functions (Wise, 1992; Corey, 1994). In soil physics literature, van Genuchten (1980) and Brooks and Corey (1964) relations are the most commonly used soil-water retention functions. Descriptions of porous media properties based on both of these functions are considered in this study.

Models Based on the van Genuchten Function

To describe water retention characteristics of soils, van Genuchten (1980) proposed an empirical relationship between relative water saturation and pressure head:

\[
\Theta = \left( \frac{1}{1 + [\alpha \psi]^{n_v}} \right)^{m_v} \]

where \( \Theta \) is the relative saturation, \( \psi \) is the absolute value of the capillary pressure head (L), \( n_v \) is the PSD index, \( \alpha \) is a parameter whose inverse is referred to as the air entry value or bubbling pressure, and \( m_v \) is normally assumed to be equal to \( 1 - 1/n_v \). Relative saturation \( \Theta \) is defined as

\[
\Theta = \frac{\theta - \theta_r}{\theta_s - \theta_r}
\]

where \( \theta \) is the water content, \( \theta_r \) is the residual water content (here taken as zero), and \( \theta_s \) is saturated water content (here assumed to be equal to the soil porosity). The pressure head \( \psi \) is related to pore radius through the capillary law, \( r = C/\psi \), and the capillary constant \( C \) is expressed as

\[
C = \frac{2\sigma \cos \phi}{\rho g}
\]

where \( \sigma \) is the interfacial tension between air and water phase (MT⁻²), \( \phi \) is the contact angle, \( \rho \) is the density of water (ML⁻³), and \( g \) is acceleration due to gravity (LT⁻²). According to the van Genuchten model, porous media with a wide range of pores have small \( n_v \) values while those with uniform pores have large \( n_v \). Grain-size distribution has little effect on \( n_v \); any type of soil, regardless of its grain-size distribution, can be made to have a uniform pore-size distribution by proper mixing and packing (Corey, 1994).

The PSD function can be derived from the soil-water retention function through the differential relation, \( f(\tau) = d\Theta/d\tau \) (Wise et al., 1994). Differentiating equation (3) after incorporating the capillary law, an expression for the PSD function can be written as (Wise et al., 1994),

\[
\frac{f(\tau)}{n_o} = m_v n_v (\alpha, C) n_v \tau^{-(n_v + 1)} \left[ 1 + (\alpha, C) n_v \tau^{n_v - 1} \right]^{-(m_v + 1)}
\]

The PSD function (6), derived from the van Genuchten function, accounts for all possible pore radii between zero and infinity. A detailed analysis of equation (6) is given in Wise et al. (1994).

Substituting the PSD function into equations (1) and (2) and integrating the resulting expressions between limits zero to infinity gives the following equations for permeability and specific surface area for a clean porous medium:

\[
\overline{k}_o = \tau G n_o \sqrt{\frac{1}{1 + [\alpha \psi]^{n_v}}}
\]

\[
\overline{M}_o = 2 \frac{n_v (n_v - 1)}{\alpha_v n_v} B(z, w)
\]

where \( B \) is the complete beta function defined by the integral (e.g., Press et al., 1992)

\[
B(z, w) = \int_0^1 t^{z-1} (1 - t)^{w-1} \, dt
\]

where \( z = (-2/n_v + 1) \), \( w = (1/n_v + 1) \), overbar identifies the variables derived from the van Genuchten model, and index "o" is used to represent a clean porous medium.

The form of the permeability model [equation (7)] is similar to that of the saturated hydraulic conductivity model presented in Mishra and Parker (1990). From equation (7), one could infer that the absolute permeability value derived from van Genuchten's model is independent of \( n_v \). This insensitivity is due to the commonly used assumption that \( m_v = 1 - 1/n_v \) (Mishra and Parker, 1991), the validity of which is discussed in van Genuchten and Nielsen (1985).

Since inverse of \( \alpha \) is the entry pressure (van Genuchten and Nielsen, 1985), from the capillary law the product \( \alpha C \) reduces to \( R \) (maximum pore radius). Using this relation, equations (7) and (8) are further simplified as

\[
\overline{k}_o = \tau G n_o R^2
\]

\[
\overline{M}_o = \frac{2n_v (n_v - 1)}{Rn_v} B(z, w)
\]

Equations (10) and (11) can be used to estimate the permeability and specific surface area of clean porous media.
Macroscopic Models for Biomass-Affected Porous Media Properties

The macroscopic approach used for modeling porous media biomass is similar to those used by Borden and Bedient (1986), Kindred and Celia (1989), and “option A” of Baveye and Valocchi (1989). Microorganisms are assumed to exist in both aqueous and solid phases, and biomass growth and nutrient consumption occur in both phases. Changes in porous media properties are assumed to be caused by accumulation of solid-phase biomass in pore spaces. The fraction of volume occupied by the solid-phase biomass can be estimated as

\[
n^f = \frac{X^f \rho_k}{\rho_t}
\]

(12)

where \(n^f\) is the volume fraction of the biomass (L\(^3\) biomass/L\(^3\) total), \(X^f\) is the mass (dry weight) of microbial cells per unit mass of aquifer solids, \(\rho_k\) is the bulk density of aquifer solids (ML\(^{-3}\)), and \(\rho_t\) is the solid-phase biomass density (ML\(^{-3}\)).

Based on volume balances, a simple macroscopic equation for computing porosity reduction can be written as

\[
n_b = n_o - n^f
\]

(13)

where \(n_b\) is the biomass-affected porosity. Using this porosity relationship, equations (10) and (11) are modified to estimate biomass-affected permeability \(k_s\) and specific surface area \(M_b\) values,

\[
\frac{k_b}{k_o} = \left(1 - \frac{n^f}{n_o}\right)^{5/2} \left(\frac{R_b}{R}\right)^2
\]

(14)

\[
\frac{M_b}{M_o} = \frac{n_b R}{n_o R_b}
\]

(15)

where index “b” is used to represent biomass-affect ed porous media properties.

In extending equation (11) to biomass-affect ed porous media [equation (15)], we ignore changes in PSD caused by biological growth and account only for reductions in the maximum pore radius. This assumption is made based on the experimental results presented by Torbati et al. (1986), who studied the in situ growth of microorganisms in Berea sandstone cores and observed that microbes preferentially plug larger pores having high nutrient flow. Permeability reductions were primarily caused by microbial growth in the large pores. They observed some increases in the frequency of small pores and decreases in the frequency of large pores, but no significant changes in the overall width of the pore-size distributions.

Dividing equations (10) and (11) by equations (14) and (15), respectively, yields expressions for relative changes in porous media properties due to biomass accumulation,

\[
\frac{k_b}{k_o} = \left(1 - \frac{n^f}{n_o}\right)^{5/2} \left(\frac{R_b}{R}\right)^2
\]

(16)

\[
\frac{M_b}{M_o} = \frac{n_b R}{n_o R_b}
\]

(17)

As a porous medium accumulates biomass, porosity will decrease and the entry pressure will increase. Relative changes in the entry pressure of biomass-affect ed porous media should be proportional to changes in porosity values. Since entry pressure is inversely related to the maximum pore radius through the capillary relation, we assume the following expression for modeling \(R_b\),

\[
\left[\frac{R_b}{R}\right]^m = \frac{n_b}{n_o}
\]

(18)

where \(m\) is an empirical constant. Substituting the capillary law into equation (18) gives,

\[
\left[\frac{\psi^e}{\psi^c}\right]^m = \left[\frac{C}{C_b}\right]^m \frac{n_b}{n_o}
\]

(19)

where \(\psi^e\) is the entry pressure head of clean porous media, \(\psi^c\) is the entry pressure head of biomass-affect ed porous media, and \(C\) is the biomass-affect ed capillary constant. Equation (19) shows a possible proportionality relationship between entry pressure and porosity variations, with \((C/C_b)^m\) being the proportionality constant.

A value for empirical constant \(m\) can be estimated by assuming that porous media consist of cylindrical pores of effective lengths proportional to radii. This simple assumption for pore structure is commonly used by several other researchers for modeling porous media properties (Fatt, 1956; Mualem, 1976; Mishra et al., 1989; Wise, 1992). Moreover, photomicrographic studies of porous media also show that the lengths of pores are proportional to their radii (Dullien, 1979). Using this assumption, a ratio between clean and biomass-affect ed pore volumes would yield an expression similar to equation (18) for the value of \(m = 3\). Through sensitivity analyses, other possible values for \(m\) were also explored and \(m = 3\) was found to be the most appropriate value. The details of this analysis are discussed in the results section.

Using (18) with \(m = 3\), the equations for relative changes in permeability (16) and specific surface area (17) can be simplified to

\[
\frac{k_b}{k_o} = \left(1 - \frac{n^f}{n_o}\right)^{19/5}
\]

(20)

\[
\frac{M_b}{M_o} = \left(1 - \frac{n^f}{n_o}\right)^{2/3}
\]

(21)

The macroscopic model equations (13), (20), and (21) together with equation (12) can be used to compute biomass-affect ed porosity, specific surface area, and permeability values at any attached biomass concentration.

Models Based on the Brooks and Corey Function

Brooks and Corey (1964) proposed an empirical equation to describe the relationship between relative water saturation and capillary pressure head,

\[
\theta = \left(\frac{\psi^c}{\psi}\right)^\lambda
\]

(22)

where \(\psi^c\) is the entry or bubbling pressure head (L), and \(\lambda\) is a PSD index similar to the van Genuchten parameter \(n\). Using the capillary law, equation (22) can be modified as:

\[
\theta = \left(\frac{r}{R}\right)^\lambda
\]

(23)

where \(r\) and \(R\) are pore radii corresponding to the pressure head \(\psi\) and \(\psi^c\), respectively. By differentiating (23), an expression for \(f(r)\) can be written as
where the constant $\beta_o$ is equal to $\lambda n_o$. This PSD function accounts for pore sizes up to a radius $R$ that corresponds to the entry pressure head. Equation (24) is the same as the power function for $f(r)$ assumed by Taylor et al. (1990). In order to compare macroscopic models against Taylor et al.’s (1990) biofilm models, the macroscopic expressions are briefly re-derived from equation (24). Incorporating (24) into (1) and (2) and integrating the resulting expressions between the limits $r = 0$ to $R$, we obtain

$$\tilde{\kappa}_o = \frac{\tau G \lambda^2 R^2 n_o^{5/2}}{(1 + \lambda)^2}$$

$$\tilde{M}_o = \frac{2\lambda n_o}{R(\lambda - 1)}$$

where variables denoted with a tilde are derived from the Brooks-Corey model. To estimate biomass-affected specific surface area and permeability values, equations (25) and (26) are modified as:

$$\tilde{\kappa}_b = \frac{\tau G \lambda^2 R^2 \tilde{n}_b^{5/2}}{(1 + \lambda)^2}$$

$$\tilde{M}_b = \frac{2\lambda \tilde{n}_b}{R_b(\lambda - 1)}$$

where $R_b$ is the biomass-affected maximum pore radius. Similar to the previous section, equations (27) and (28) are written after ignoring variations in PSD index values due to biological growth. Dividing (27) by (25) and (28) by (26), respectively, and invoking the relation between $R$ and $R_b$ from equation (18) and the porosity relation from equation (13) yields

$$\frac{\tilde{\kappa}_b}{\tilde{\kappa}_o} = \left(1 - \frac{n'}{n_o}\right)^{19/6}$$

$$\frac{\tilde{M}_b}{\tilde{M}_o} = \left(1 - \frac{n'}{n_o}\right)^{2/3}$$

The relative permeability and specific surface area model equations (29) and (30) are exactly the same as equations (20) and (21). This analysis shows that, under the assumed conditions, macroscopic model-based expressions for relative permeability and specific surface area derived from van Genuchten and Brooks-Corey functions are identical.

**Results and Discussion**

**Comparison of Macroscopic Models to Biofilm-Based Models**

A comparison of results based on our macroscopic models and those of Taylor et al. (1990) biofilm models is presented for predictions of relative changes in porosity, permeability, and specific surface area. Porous media parameters used for model comparison are: $n_o = 0.347, \tau_o = 0 \text{ cm}, \tau = 0.903$, and $R = 0.0286 \text{ cm}$ (Taylor and Jaffé, 1990c). However, all simulation results will be summarized in a normalized form and are not sensitive to the choice particular values for these parameters. The value for $\lambda$ was varied within the range of $2 \leq \lambda \leq 5$, typical for aquifers with sandy porous media (Corey, 1994).

Simulation experiments started with clean porous media and accumulated an incremental amount of biomass $dn'$ at every discrete computational step. In all simulations $n'/n_o$ was treated as the independent variable. For the macroscopic approach, changes in porous media properties were directly computed from equations (13), (20), and (21).

In their biofilm approach, Taylor and Jaffé (1990c) use the following approximate expression to relate biomass fraction ($n'$), biofilm length ($L_f$), and specific surface area ($M$) values,

$$n' = M_0 L_f$$

Because specific surface area of a biologically active porous medium is a function of $L_f$, we use a differential form of this expression, which is a more rigorous and appropriate method, to relate the three variables,

$$\frac{dL_f}{dn'} = \frac{1}{M_0 (L_f)}$$

An analytical expression for $M_0 (L_f)$ was given by Taylor et al. (1990) (refer to the Appendix). A predictor-corrector Euler approximation was used to solve equation (32) for $L_f$, and the biofilm lengths were later used to compute biofilm-affected porous media properties $n_o (L_f)$ and $k_o (L_f)$ (refer to the Appendix for Taylor et al. (1990) equations). Other numerical integrals in Taylor et al. (1990) models were evaluated using the trapezoidal rule.

Figure 1 presents the porosity values predicted by both biofilm and macroscopic models. Since the macroscopic model for porosity is an exact statement of volume balance, the biofilm model-based porosity values should relax to macroscopic values regardless of porous media characteristics. This result is apparent in the predicted porosity profiles presented in the figure.

Figure 2 shows permeability reductions predicted by both the models. It can be seen from the figure that biofilm approach results corresponding to $\lambda = 3$ are in exact agreement with macroscopic model predictions. Moreover, in the range of $2 \leq \lambda \leq 5$, the relative permeability profiles predicted by the biofilm-based model are fairly insensitive to $\lambda$ and closely follow those obtained by the macroscopic approach. The maximum absolute deviation in the relative permeability values is approximately 3% of the initial permeability value. A precise agreement between macroscopic and biofilm model predictions at $\lambda = 3$ can be established analytically. The Appendix contains pertinent mathematical derivations that prove that at $\lambda = 3$, the porosity,
specific surface area, and permeability equations of Taylor et al. (1990) exactly relax to the macroscopic equations.

In Figure 3, the values of specific surface area computed by the two approaches are compared. Similar to the permeability profiles, both models match exactly at $\lambda = 3$. However, at other $\lambda$ values, there are some discrepancies between model predictions. An attempt was made to reduce these discrepancies by varying the constant $m$ in equation (18) as a function of $\lambda$. The following analysis is purely empirical and assumes that Taylor et al. (1990) models are accurate. By changing $m$ values, the macroscopic model predictions are simply tuned to fit Taylor et al.'s (1990) model predictions.

Using equation (18) in the general form, macroscopic expressions for specific surface area and permeability are modified as

$$\frac{M_b}{M_o} = \left[ 1 - \frac{n^{(1/m)}}{n_o} \right]^{1-(1/m)} \quad (33)$$

$$\frac{k_b}{k_o} = \left[ 1 - \frac{n^{(5/2)+(1/m)}}{n_o} \right] \quad (34)$$

Figure 4 depicts the sensitivity of equations (33) and (34) to variations in $m$. Results show that a close agreement between the macroscopic and biofilm models is achieved at $\lambda = 5$ and $m \approx 2.2$, and at $\lambda = 2.0$ and $m = 20$; thus $m$ is a relatively insensitive parameter, and use of $m = 3$ is a reasonable approximation for uniform porous media.

Values of specific surface area are typically unimportant except in the biofilm approach where they are required to compute film thicknesses. Although the macroscopic approach does not explicitly consider biofilms, it is possible to estimate apparent biofilm thicknesses through macroscopic model-based equations (32) and (21). These film thicknesses can be useful in modeling diffusional limitations across attached biomass (Rittmann and McCarty, 1981). For the example problem, the maximum deviation between specific surface area values seen in Figure 3 is about 10% of the initial specific surface area. This approximately transforms into 10% error in biofilm-length estimates, which is relatively minor given the uncertainties in values of biofilm density, maximum pore radius, and other soil/biofilm parameters. Hence, macroscopic models with $m = 3$ may also be used for modeling reactive transport under diffusion-limited conditions.

**Comparison Based on a Reactive Ground-Water Transport Model**

To further compare the macroscopic and biofilm approaches, a lab-scale, reactive transport experiment reported in Zysset et al. (1994) is simulated. The experiment involves transport of a limiting nutrient (nitrate), and growth and decay of both attached- and aqueous-phase biomass in a 10 cm column. Zysset et al. (1994) modeled the experimental data with a reactive transport model that does not consider porosity variations. For

Fig. 4. Comparison of specific surface area and relative permeability profiles predicted by the biofilm model at different "$\lambda$" values and the macroscopic model at different "$m$" values.
modeling purposes, they expressed attached biomass concentrations \((X')\) as grams of biomass per liquid volume \((\text{ML}^{-3}\text{T}^{-1})\). Here, to be consistent with our previous definitions, we express these concentrations as grams of biomass per unit mass (dry bulk mass) of porous media solids \((\text{MM}^{-1}\text{T}^{-1})\). With this modification, the governing transport equations for the limiting nutrient and suspended biomass are written as

\[
\frac{\partial (nC_w)}{\partial t} = q \alpha \frac{\partial^2 C_w}{\partial x^2} - q \frac{\partial C_w}{\partial x} + k_w \left( \frac{X_{\text{max}} - X'}{X_{\text{max}}} \right) C_w X' \rho_k - k_m \left( \frac{C_w}{\delta + C_w} \right) X' \rho_k \quad (35)
\]

\[
\frac{\partial (nC_b)}{\partial t} = q \alpha \frac{\partial^2 C_b}{\partial x^2} - q \frac{\partial C_b}{\partial x} - n k_s C_s + k_s X' \rho_k \quad (36)
\]

Growth and substrate utilization of attached phase biomass are modeled using the differential equation

\[
\frac{dX'}{dt} = u_f k_s \left( \frac{X_{\text{max}} - X'}{X_{\text{max}}} \right) C_w X' - (k_d + k_s) X' + \frac{n k_s C_s}{\rho_k} \quad (37)
\]

where \(n\) is the porosity, \(q\) is the Darcy flux \((\text{LT}^{-1})\), \(C_w\) is the water-phase substrate concentration of the limiting substrate \((\text{ML}^{-3})\), \(C_b\) is the water-phase concentration of biomass \((\text{ML}^{-3})\), and \(X'\) (similar to the variable \(C_b\) used by Zysset et al., 1994) represents the attached biomass concentration \((\text{MM}^{-1})\), \(\alpha\) is the dispersivity \((\text{L})\), \(\delta\) is a switching parameter, \(k_s\) is the specific growth rate \((\text{M}^{-1}\text{L}^{-3}\text{T}^{-1})\), \(k_m\) is the maintenance rate \((\text{T}^{-1})\), \(k_a\) is the adsorption rate \((\text{T}^{-1})\), \(k_d\) is the desorption rate \((\text{T}^{-1})\), \(k_d\) is the decay rate \((\text{T}^{-1})\), and \(u_f\) is the stoichiometric coefficient. We also converted the value of \(X_{\text{max}}\) used in Zysset et al. (1994) into a unit consistent with \(X'\) values using a relation \(X_{\text{max}} = n a X_{\text{zymax}}/\rho_k\). A summary of values for kinetic and column parameters used in these simulations is given in Table 1.

Soil parameters are identical in value to those used in the previous section. The soil column was initially assumed to be clean, and feed solution containing \(1.5 \times 10^{-6} \text{ g/l}\) of biomass was used to inoculate the soil for 0.1 days. The feed concentration of nitrate was maintained continuously at a concentration of \(5.5 \times 10^{-6} \text{ mol/l}\).

**Table 1. Summary of Model Parameters**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>(q)</td>
<td>1.833 m d(^{-1})</td>
</tr>
<tr>
<td>(\alpha)</td>
<td>0.5 m</td>
</tr>
<tr>
<td>(n_0)</td>
<td>0.39</td>
</tr>
<tr>
<td>(\rho_t)</td>
<td>2.5 g l(^{-1})</td>
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<tr>
<td>(\rho_b)</td>
<td>1600 g l(^{-1})</td>
</tr>
<tr>
<td>(k_w)</td>
<td>225 g l(^{-1}) d(^{-1})</td>
</tr>
<tr>
<td>(k_m)</td>
<td>0.05 mol g(^{-1}) d(^{-1})</td>
</tr>
<tr>
<td>(k_a)</td>
<td>50 d(^{-1})</td>
</tr>
<tr>
<td>(k_d)</td>
<td>0.0</td>
</tr>
<tr>
<td>(k_s)</td>
<td>0.05 d(^{-1})</td>
</tr>
<tr>
<td>(u_f)</td>
<td>7.5 g mol(^{-1})</td>
</tr>
</tbody>
</table>
| \(X_{\text{max}}\) | 2.925 \times 10^{-6} \text{ g} \text{g}^{-1}\)

[Reaction constants are from Zysset et al. (1994).]

The coupled transport equations (35) and (36) were solved by an iterative, finite-difference procedure with Picard linearization for the nonlinear terms (Molz et al., 1986). The reaction equation (37) was solved at each Picard level, by the fourth-order Runge-Kutta method. To achieve a convergent solution, approximately three Picard iterations were required within each time step. Uniform spatial grids 0.2 cm in length and 0.01 hr in time were used. The code was first run under constant porosity conditions to validate the model predictions against the results reported by Zysset et al. (1994). The model results were also verified for numerical correctness through mass balance computations of the transported species.

To compare the results and performance of macroscopic and biofilm-based reactive transport models, simulations were run under variable porosity conditions. Under similar computing conditions, the macroscopic-model-based transport code ran approximately 25 times faster than the biofilm-model-based code. Transient variations in porosity and permeability values predicted by both models are reported in Figures 5 and 6. Although variations in porosity and permeability values in the column are appreciable, differences between the profiles predicted by biofilm and macroscopic models are negligible. The porosity profiles are identical, and the relative permeability profiles have only small deviations. At the column entrance, a maximum absolute relative permeability deviation of 2.5% of the initial value is observed.
In spite of substantial changes in porosity values within the column, nutrient concentration profiles predicted by the numerical model under variable and constant porosity conditions were nearly the same (results not shown). This indicates that the biological reaction rates in Zysset et al.’s (1994) column are rather rapid and hence reduced nutrient retention times due to porosity reductions did not affect the amount of nutrient consumption. This observation shows that for rapidly reactive systems, indirect evidences such as variations in breakthrough nutrient profiles, may not be sufficiently sensitive to infer changes in porous media properties.

Summary and Conclusions

Analytical models for predicting changes in porosity, specific surface area, and permeability due to microbial growth in porous media are developed from a macroscopic approach. Variations in soil properties are computed from macroscopic-averaged attached biomass concentrations. The macroscopic description of biomass is conceptually in better agreement with the Darcy-scale continuum models.

The macroscopic models developed in this work are easy to implement and computationally more efficient than the biofilm models. The macroscopic expressions can be explicitly used to estimate biomass-affected porosity, specific surface area, and permeability at any attached biomass concentration. Since the biofilm models of Taylor et al. (1990) are implicit functions of attached biomass thickness, these porous media properties cannot be estimated explicitly. For a porous medium with a value of \( \lambda = 3 \), macroscopic model predictions of porosity, specific surface area, and permeability changes are in exact agreement with the biofilm model predictions. At other values of \( \lambda \) (2 \( \leq \lambda \leq 5 \)), simulated porosity profiles are almost identical, relative specific surface area and permeability profiles have maximum absolute deviations of 10% and 3% of their respective initial values.

Column simulations show that under similar computing conditions, macroscopic approach-based transport models are computationally more efficient than biofilm approach-based transport models. Differences in the porosity and permeability profiles predicted by macroscopic and biofilm approaches are almost negligible showing a maximum absolute deviation of 2.5% of the initial value. The proposed models can be used as simple tools for estimating changes in porous media properties during bioremediation of contaminated aquifers.

Appendix

Analytical Comparison of Macroscopic and Taylor et al. (1990) Biofilm-Based Models

Taylor et al. (1990) used a cut-and-random-rejoin-type model to describe changes in the physical properties of a biofilm-coated porous medium. They model pore-size distributions with a power function similar to the Brooks-Corey PSD function. Their analysis initially assumes a clean porous medium described by fixed values of \( n_o, \lambda, \beta, r_o, \) and \( R \). Changes in the porous medium properties due to biological growth are computed incrementally by assuming a uniform, impermeable biofilm of thickness \( dL_f \). Other than the conceptual differences between biofilm and macroscopic models, a distinct difference between present and Taylor et al. derivations is that in Taylor et al.’s work the parameters \( \beta \) and \( R \) are treated as constants, whereas, in the present work they are treated as variables.

The analytical models presented by Taylor et al. (1990) for predicting clean and biofilm-affected porosity, specific surface area, and permeability values are

\[
\hat{n}_o = \frac{\beta}{\lambda} \left[ 1 - \left( \frac{r_o}{R} \right)^{\lambda} \right] \tag{38}
\]

\[
\hat{M}_s = \frac{2\beta}{R(\lambda - 1)} \left[ 1 - \left( \frac{r_o}{R} \right)^{\lambda - 1} \right] \tag{39}
\]

\[
\hat{k}_o = \frac{\tau \beta^2 R^2 n_o^{1/2}}{8} \left[ \frac{1}{1 + \lambda} - \left( \frac{r_o}{R} \right)^{1+\lambda} \right]^2 \tag{40}
\]

\[
\hat{n}_b = \beta \left( \frac{L_f}{R} \right)^\lambda \left[ I_2 \left( \frac{R}{L_f} - 1, \lambda \right) - I_2 \left( \frac{r_{ob}}{L_f}, \lambda \right) \right] \tag{41}
\]

\[
\hat{M}_b = \frac{2\beta}{L_f} \left( \frac{L_f}{R} \right)^\lambda \left[ I_1 \left( \frac{R}{L_f} - 1, \lambda \right) - I_1 \left( \frac{r_{ob}}{L_f}, \lambda \right) \right] \tag{42}
\]

\[
\hat{k}_b = \frac{\tau n_b^{1/2} \beta^2 L_f^2}{8} \left( \frac{L_f}{R} \right)^{2\lambda} \left[ I_1 \left( \frac{R}{L_f} - 1, \lambda \right) - I_1 \left( \frac{r_{ob}}{L_f}, \lambda \right) \right]^2 \tag{43}
\]

where the symbol \(^\wedge\) (hat) is used to represent variables based on the biofilm approach, \( L_f \) is the biofilm thickness, and \( r_{ob} = \max(r_o - L_f, 0) \). Values of \( \hat{n}_o \) and \( n_o \) are identical, since both of them represent initial porosity of clean porous media. Functions \( I_1, I_2, \) and \( I_3 \) are defined as:

\[
I_n(u, \lambda) = \int_0^u \frac{x^n}{(1 + x)^{3+\lambda}} \, dx \tag{44}
\]

where \( n = 1, 2, \) or 3 and \( u = R/L_f - 1 \) or \( r_{ob}/L_f \).

For comparison purposes, a porous medium with values of \( \lambda = 3 \) and \( r_o = 0 \) is assumed in the following analysis. Under the assumed conditions, the integral (44) reduces to a simple form

\[
I_n(u, 3) = \frac{u^{n+1}}{n+1} \tag{45}
\]

Using (45), specific surface area model equations (39) and (42) can be simplified as,

\[
\hat{M}_s = \frac{\beta}{R} \tag{46}
\]

\[
\hat{M}_b = \frac{\beta}{R} \left( \frac{u L_f}{R} \right)^2 \tag{47}
\]

Dividing (47) by (46) we get,

\[
\frac{\hat{M}_b}{\hat{M}_s} = \left( \frac{u L_f}{R} \right)^2 \tag{48}
\]

Similarly an expression for the ratio between biomass-affected and clean porosities can be derived as,

\[
\frac{\hat{n}_b}{n_o} = \left( \frac{u L_f}{R} \right)^{1/3} \tag{49}
\]

To further simplify these ratios, we incorporate specific surface area expression (47) into biofilm relation (32) after substituting for \( u \), and obtain the following differential equation,
\[
\frac{dn^f}{dL_f} = \beta \left[ \frac{L_f}{R} - \frac{2L_f}{R^2} + \frac{L_f^2}{R^3} \right] \tag{50}
\]

Solving (50) with initial conditions: \( n^f = 0 \) at \( L_f = 0 \), and \( \beta = \lambda n_o \), we obtain,

\[
1 - \frac{n^f}{n_o} = \left( \frac{uL_f}{R} \right)^3 \tag{51}
\]

From (49), equation (51) can be written in terms of porosity as

\[
\hat{n}_o = n_o - n^f \tag{52}
\]

(or from (48), equation (51) can be expressed in terms of specific surface area as

\[
\frac{\dot{M}_b}{\dot{M}_o} = \left( 1 - \frac{n^f}{n} \right)^{2/3} \tag{53}
\]

Using similar analyses, the ratio between biofilm-affected and clean porous media permeabilities can also be derived as

\[
\frac{\hat{k}_b}{\hat{k}_o} = \left( \frac{uL_f}{R} \right)^8 \left( \frac{\hat{n}_b}{n_o} \right)^{3/2} \tag{54}
\]

This expression can be simplified to

\[
\frac{\hat{k}_b}{\hat{k}_o} = \left( 1 - \frac{n^f}{n_o} \right)^{19/6} \tag{55}
\]

Equations (52), (53), and (55) derived from Taylor et al.'s (1990) biofilm-based models are identical to macroscopic model equations (13), (21), and (20), respectively. This demonstrates the agreement between biofilm and macroscopic models at \( \lambda \) equal to 3.

Further, if \( n^f/n \) is treated as relative saturation, then equation (55) is the same as the Brooks-Corey based relative permeability model derived by Mualem (1976). However, this agreement between Taylor et al.'s (1990) biofilm-affected relative permeability model and Mualem's unsaturated relative permeability model would occur only at \( \lambda = 3 \).

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Important Notations

\begin{itemize}
  \item \textbf{B} Complete beta function.
  \item \textbf{C} Capillary constant.
  \item \textbf{f(r)} Pore-size distribution function.
  \item \textbf{k} Soil permeability (L²).
  \item \textbf{G} Geometric factor.
  \item \textbf{g} Acceleration due to gravity (LT⁻²).
  \item \textbf{L_f} Biofilm length.
  \item \textbf{m} Empirical constant.
  \item \textbf{M} Specific surface area (L⁻¹).
  \item \textbf{n} Porosity.
  \item \textbf{n'} Volume fraction of the biomass (L³ biomass/L³ total).
  \item \textbf{n_v} Pore-size distribution index—van Genuchten model.
  \item \textbf{q} Darcy flux (LT⁻¹).
  \item \textbf{r} Porous radius (L).
  \item \textbf{R} Maximum pore radius (L).
  \item \textbf{X'} Mass (dry mass) of microbial cells per unit mass of aquifer solids.
  \item \textbf{\tau} Tortuosity factor.
  \item \textbf{\theta} Water content.
  \item \textbf{\theta_r} Residual water content.
  \item \textbf{\theta_s} Saturated water content.
  \item \textbf{\Theta} Relative saturation.
  \item \textbf{\psi} Capillary pressure head (L).
  \item \textbf{\psi_e} Entry pressure head of clean porous media (L).
  \item \textbf{\alpha} Dispersivity (L).
  \item \textbf{\alpha_v} van Genuchten parameter (L⁻¹).
  \item \textbf{\beta} Brooks-Corey model parameter.
  \item \textbf{\delta} Switching parameter.
  \item \textbf{\lambda} Pore-size distribution index—Brooks-Corey model.
  \item \textbf{\sigma} Interfacial tension between air and water phase (MT⁻²).
  \item \textbf{\phi} Contact angle.
  \item \textbf{\rho} Density of water (ML⁻³).
  \item \textbf{\rho_k} Bulk density of aquifer solids (ML⁻³).
  \item \textbf{\rho_i} Solid-phase biomass density (ML⁻³).
  \item \textbf{\nu} Stoichiometric coefficient.
\end{itemize}

Also, a subscript index "o" is used to represent clean porous media parameters, a subscript index "b" is used to represent biomass-affected parameters, an overbar is used to represent van Genuchten model-based variables, a tilde is used to represent Brooks-Corey model-based variables, and a hat is used to represent Taylor et al.'s (1990) biofilm model-based variables.

References


