Modeling of a microbial growth experiment with bioclogging in a two-dimensional saturated porous media flow field

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Abstract

A model was developed simulating reactive transport in groundwater including bioclogging. Results from a bioclogging experiment in a flow cell with a two-dimensional flow field were used as a data base to verify the simulation results of the model. Simulations were performed using three different hydraulic conductivity vs. porosity relations published in literature; two relations derived from pore network simulations assuming the biomass to grow in discrete colonies and as a biofilm, respectively, and a third relation, which did not include pore connectivity in more than one dimension. Best agreement with the experimental data was achieved using a hydraulic conductivity vs. porosity relation derived from pore network simulation assuming the biomass to grow in colonies. The relation derived from pore network simulations assuming biomass to grow as a biofilm was unable to reproduce the experimental data when realistic parameter values were employed. With the third relation the clogging ability of the biomass was strongly underestimated. These findings indicate that the porous medium needs to be treated as a multi-dimensional medium already on the pore scale, and that biomass growth different than in a biofilm must be considered to get an appropriate description of bioclogging.

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1. Introduction

The change in porosity and hydraulic conductivity of a saturated porous medium due to microbial growth is commonly referred to as bioclogging (Baveye et al., 1998). Previous investigations have demonstrated that bioclogging may influence the success of bioremediation in aquifers (Anderson and Lovley, 1997), the performance of sand filters (Urfer et al., 1997), or the effectiveness of water flooding of oil reservoirs (Lappan and Fogler, 1994).

Thus, it is important to understand the mechanisms involved in bioclogging. Most of the studies on bioclogging were performed in laboratory systems with one-dimensional flow fields (e.g., Cunningham et al., 1991; Taylor and Jaffé, 1990; Vandevivere and Baveye, 1992). These authors observed high reduction in hydraulic conductivity (two to three orders of magnitude) due to biomass production. Microbial growth and its influence on hydraulic properties of microscopic pore networks were investigated by Dupin and McCarty (2000) and Kim and Fogler (2000). Both observed a decrease in hydraulic conductivity (two orders of magnitude) and Dupin and McCarty (2000) also reported on a correlation between growth conditions and the morphology of bacterial growth (e.g., formation of colonies or biofilms depending on pH). Recently, Kildsgaard and Engesgaard (2002) and Thullner et al. (2002a) investigated bioclogging in systems with two-dimensional flow fields. In these systems bioclogging was also observed.

However, in contrast to one-dimensional systems, effects like flow bypass played an important role in the two-dimensional flow fields. Whereas no measurements of the biomass were performed by Kildsgaard and Engesgaard (2002), Thullner et al. (2002a) monitored the distribution and quantified the amount of biomass in the porous medium, showing that biomass mainly consisted of extracellular polymeric substances (EPS). This demonstrated that due to EPS production a relatively small amount of bacteria can be responsible for significant clogging effects.

Several models have been introduced to simulate the experimentally observed interaction between biomass growth and changes in hydraulic properties of porous media. Usually, these models are based on the assumption of a homogeneous biofilm covering the surface of the grains (Taylor et al., 1990), and a reduction of hydraulic conductivity caused by biofilm growth is theoretically derived assuming bundles of parallel pores. Especially for fine-textured materials these models were unable to satisfactorily predict observed hydraulic conductivity reductions, which was attributed to microorganisms growing in colonies as compared to growth in a biofilm (Vandevivere, 1995; Vandevivere et al., 1995). In addition, it was suggested to use pore networks instead of pore bundles in order to account for interpore connections (Loehle and Johnson, 1994; Vandevivere et al., 1995). Clement et al. (1996) presented a macroscopic generalization of the approach of Taylor et al. (1990), but their prediction of reductions in hydraulic conductivity was nearly identical to the biofilm model of Taylor et al. (1990). A pore network model has been introduced and applied to previously published data (Suchomel et al., 1998a,b). The model produced realistic results assuming the growth of a biofilm on the walls of cylindrical pores. In contrast to this, Dupin et al. (2001) indicated that the experimental observations of Dupin and McCarty (2000) could best be explained with network model simulations assuming the growth of biomass in aggregates, whereas assuming a biofilm could not explain their observations. However,
they did not present a functional relation between reductions in hydraulic conductivity and porosity.

In a recent study, Thullner et al. (2002b) used pore network models to investigate the influence of biomass on hydraulic conductivity. Two different hydraulic conductivity vs. porosity relations were presented (in the following referred to as $k_f-n$ relations), which predict the decrease of hydraulic conductivity caused by porosity reductions, depending on the morphology of microbial growth. Both relations predict a higher reduction of hydraulic conductivity for a given reduction of porosity than the relations published previously (e.g., Clement et al., 1996).

![Diagram of the experimental design](image-url)

**Fig. 1.** Diagram of the experimental design as given in Thullner et al. (2002a) (only flow cell and objects inside flow cell are drawn to scale).
In the present study, the reactive transport groundwater model TBC (transport, biochemistry, and chemistry) by Schäfer et al. (1998a) was used as the basis for developing the model TBCC (transport, biochemistry, chemistry, and clogging), which now includes bioclogging. TBCC was used to simulate the two-dimensional experimental results from Thullner et al. (2002a). Using different $k_f - n$ relations (Clement et al., 1996; Thullner et al., 2002b), we wanted to investigate whether the results of the experiment (i.e., measured biomass concentration and observed changes of the flow field due to bioclogging) could be reproduced by the model in general, and whether $k_f - n$ relationships derived from pore network models were able to predict experimental results better than $k_f - n$ relations derived from models not accounting for interpore connections.

2. Review of the two-dimensional bioclogging experiment

A bioclogging experiment was performed in a flow cell ($56 \times 44 \times 1$ cm, Fig. 1) filled with a water-saturated glass bead packing and operated under a continuous flow of a mineral medium containing nitrate (injected via four inflow ports at the bottom of the flow cell, creating a two-dimensional flow field) (Thullner et al., 2002a). Simultaneously, a glucose solution was injected through an injection port, simulating a point source of carbon substrate. Visible light transmission was used to observe qualitatively the distribution of the growing biomass and water flow during the experiment. At the end of the experiment, porous medium samples were destructively collected along several transects and analyzed for abundance of bacterial cells, bacterial cell volume, and concentration of polysaccharides and proteins. Microbial growth was observed in a stripe along the length of the flow cell, starting at the glucose injection port, where the highest biomass concentrations were obtained. During a tracer test (performed at days 28 and 29 of the experiment), dyed water was injected at a tracer injection port approximately 4 cm below the position of the glucose injection port. Biofouling effects became evident when the injection of the glucose solution was turned off and water flow still bypassed the area around the glucose injection port, preserving the water flow pattern as it was during the injection of the glucose solution. Analyses of the porous media samples showed that only 5% of the total organic carbon was present as bacterial biomass, whereas the remaining 95% were attributed to EPS. The total volume of the bacterial cells remained below 0.01% of the pore space even in the vicinity of the injection port. Therefore, the observed clogging effects were assumed to be mainly caused by EPS.

3. Model description

3.1. General concept

The model TBCC (transport, biochemistry, chemistry, and clogging) used in this study is based on the reactive transport model TBC (transport, biochemistry, and chemistry) by Schäfer et al. (1998a). TBC was successfully used for simulations of a laboratory column study on microbial carbon degradation (Schäfer et al., 1998b) as well as a field experiment.
on bioremediation of a contaminated aquifer (Thullner and Schäfer, 1999). This numerical model uses a REV approach with finite element approximation to simulate three-dimensional saturated groundwater flow and transport following

\[
\nabla \cdot (k_t \nabla h) = S_0 \frac{\partial h}{\partial t} - w
\]

for water flow and

\[
\frac{\partial c_{\text{mob}}}{\partial t} = -\nabla \cdot (\overrightarrow{u} c_{\text{mob}}) + \nabla \cdot (D \nabla c_{\text{mob}}) + r
\]

for solute transport in water, where \(S_0\) is the storage coefficient and \(w\) is a source and sink term; \(D\) is the dispersion tensor including molecular diffusion. The transport velocity \(\overrightarrow{u}\) can be calculated using \(\overrightarrow{u} = \frac{\overrightarrow{v} D}{n}\), where \(\overrightarrow{v} D\) is the Darcy velocity and \(n\) the porosity, the latter is assumed to be equal to the volume of the free water phase. The term \(r\) includes all sources and sinks caused by, e.g., wells, phase exchange processes (e.g., sorption), and reactive processes (i.e., microbial consumption or chemical reactions). The piezometric head \(h\) and the solute concentration in water \(c_{\text{mob}}\) were the variables to be calculated.

In TBC the volume of each finite element is divided into three phases, the water phase (where solute transport takes place), the solid matrix phase, and a biophase. The biophase is assumed to include all microorganisms and EPS. All biological processes, especially the degradation of organic carbon, are assumed to take place in this biophase. Within each phase in a model cell, species are assumed to be distributed homogeneously. The exchange of solutes between the water phase and the biophase is simulated using

\[
\frac{\partial c_{\text{bio}}}{\partial t} = -\frac{\lambda}{n_{\text{bio}}} (c_{\text{bio}} - c_{\text{mob}})
\]

and

\[
\frac{\partial c_{\text{mob}}}{\partial t} = \frac{\lambda}{n} (c_{\text{bio}} - c_{\text{mob}})
\]

where \(\lambda\) is an exchange rate parameter. Concentration of a solute in the water phase and in the biophase is \(c_{\text{mob}}\) and \(c_{\text{bio}}\), respectively. The specific volume of the biophase is \(n_{\text{bio}}\).

Microbial growth is assumed to follow Monod-type kinetics and thus

\[
\frac{\partial X}{\partial t} = \mu_{\text{max}} X \prod_i \frac{c_{\text{bioi}}}{K_i + c_{\text{bioi}}} - \mu_{\text{dec}} X
\]

is used to express changes in bacterial mass \(X\). The Monod constants for the growth limiting solutes are \(K_i\), and \(c_{\text{bioi}}\), their concentrations in the biophase. The rate constants for maximum growth and first-order decay are \(\mu_{\text{max}}\) and \(\mu_{\text{dec}}\), respectively. Substrate consumption and release of metabolic products are coupled to microbial growth via yield coefficients and stoichiometric relations.

In order to simulate bioclogging effects, the model TBC was modified to obtain the new model TBCC. Keeping the REV approach changes in porosity are calculated by converting biomass into biovolume, which directly reduces the porosity. The hydraulic conductivity \(k_t\) is assumed to be a function of porosity \(n\). In this paper three different
relations between hydraulic conductivity and porosity are used: the macroscopic approach from Clement et al. (1996), and the pore network approach from Thullner et al. (2002b) for colony growth and biofilm growth, respectively. In Clement et al. (1996), the macroscopic model relates the change of relative hydraulic conductivity \( k_{f_{rel}} = k_f/k_{f_{ini}} \) to the change of relative porosity \( n_{rel} = n/n_{ini} \) (\( k_{f_{ini}} \) and \( n_{ini} \) are the values for the “clean” porous media (without biomass or with negligible amounts of biomass, respectively); i.e., \( n_{ini} = n + n_{bio} \)). According to Clement et al. (1996), \( k_{f_{rel}} \) is given by the function

\[
k_{f_{rel}} (n_{rel}) = n_{rel}^{19/6}
\]

(6)

In case of biomass growing in colonies, which occupy pores entirely, changes in \( k_{f_{rel}} \) are given by (Thullner et al., 2002b)

\[
k_{f_{rel}} (n_{rel}) = a \left( \frac{n_{rel} - n_0}{1 - n_0} \right)^3 + (1 - a) \left( \frac{n_{rel} - n_0}{1 - n_0} \right)^2
\]

(7)

where \( n_0 \) and \( a \) are adjustable parameters. In case of biomass growing as a homogeneous biofilm, the changes in \( k_{f_{rel}} \) are given by the function

\[
k_{f_{rel}} (n_{rel}) = \left( \frac{n_{rel} - n_0}{1 - n_0} \right)^b + k_{f_{min}} \frac{1}{1 + k_{f_{min}}}
\]

(8)

with \( n_0 \), \( k_{f_{min}} \), and \( b \) as adjustable parameters. For the latter two relations, \( 1 - n_0 \) is interpreted as the volume of biomass (relative to initial pore volume) needed to get the maximum reduction of hydraulic conductivity. In case of \( n_{rel} \leq n_0 \), it was assumed that \( k_{f_{rel}} = 0 \) in Eq. (7) or \( k_{f_{rel}} = k_{f_{min}} \) in Eq. (8), respectively. For the colony and the biofilm models, the pore network simulations predict different values for the parameters (i.e., \( n_0 \)) used in Eqs. (7) and (8) depending on the hereogeneity of the pore radius distribution. A comparison between the different models is given in Fig. 2 showing the range of possible results for the colony and biofilm model.

Using an expression from Rittmann (1982), we found that the biomass detachment rate in our experiment was several orders of magnitude smaller than the assumed biomass decay rate. For this reason, we assumed detachment to be negligible, and we did not include biomass detachment (and thus also attachment) into the model.

3.2. Implementation of model modifications

TBCC solves the flow and reactive transport problem independently for each time step, during which all flow and transport parameters are kept constant. Changes of these parameters (and all other parameters which depend on the phase volumes) due to microbial growth are calculated at the end of each simulated time step and the updated values are used for the next time step.

By giving each component of the biophase (i.e., bacteria and EPS) a specific density, the volume of the biophase is calculated for each simulated time step. The volume of the matrix phase is assumed to be constant for each element and thus the volume of the water phase or the porosity, respectively, can be calculated. As the biomass mainly consists of water
(Characklis and Marshall, 1990), it is assumed that the increasing biophase is incorporating the volume of water being removed from the water phase; a decreasing biophase is releasing water to the water phase, respectively. This procedure allows to neglect changes in hydraulic pressure due to biomass growth or decay. In addition, this exchange of water causes also an exchange of solute mass between the phases. Similar to Schäfer (1992), this mass exchange between the biophase and the water phase is calculated by assuming that, for the phase which is decreasing, the solute concentrations remain constant, and that the total solute masses in an element remain constant, too. Based on these porosity changes, the changes in hydraulic conductivity are calculated using either Eq. (6), (7), or (8).

4. Numerical simulations

4.1. General procedure

Analogous to the experiment, each simulation started on day 1 and ended on day 31. Between days 28 and 29, the glucose injection was stopped and instead a tracer solution was injected at the tracer injection port. Simulations were performed for the different $k_f-n$
relations. When using Eq. (6) the model will be referred to as Clement model, when using Eq. (7) the model will be referred to as colony model, and when using Eq. (8) the model will be referred to as biofilm model. In addition, we also performed simulations assuming no changes of hydraulic conductivity during the experiment. Apart from the differences between the $k_f-n$ relations, all parameters describing flow, transport, and reactive processes had identical values for the different simulations.

4.2. Model discretization and parameters for flow and transport

As a first preliminary step, the flow cell experiment was modeled as a whole, including the entire flow cell into the modeled area. The domain (56 cm in $x$-, 44 cm in $y$-, and 1 cm in $z$-direction) was discretized with $72 \times 65 \times 2$ nodes. The spacing in $x$- and $y$-direction ranged from 0.5 cm close to the glucose injection port to 1.0 cm towards the boundaries of the flow cell. We used a spacing of 1.1 cm in $z$-direction to address the effects of wall elasticity and the subsequent increase in thickness of the flow cell (as reported by Thullner et al. (2002a)). Positions of the inflow, outflow, and injection ports as well as injected flow rates (0.5 l/day for each inflow port and 0.04 l/day for the glucose or tracer injection port) were taken from the experiment. The outflow port was kept at a constant piezometric head of 0 m. The time step size was kept constant at 0.005 days.

For all these preliminary simulations, the biomass distribution was similar to the experimental observation of a stripe-like pattern (regardless of the used clogging model). Nevertheless, a detailed simulation of the experimental observations and especially the clogging processes was not possible with this discretization as numerical dispersion was too high. This led to an increased mixing between glucose and nitrate, and thus to a less focused distribution of biomass.

The clogging processes took place mainly in the vicinity of the glucose injection point. To obtain a detailed simulation of these clogging processes (i.e., with minimal numerical dispersion), we selected a $14 \times 56 \times 1$ cm area in the center of the flow cell. This area was modeled with a refined grid of $45 \times 84 \times 2$ nodes (Fig. 3). The spacing in $x$- and $y$-direction was 0.2 cm in an observation zone close to the glucose injection port and increased to up to 10 cm. The size of the observation zone was $8.4 \times 14.4$ cm and corresponded to the observation zone described in Thullner et al. (2002a). The spacing in $z$-direction was 1.1 cm (i.e., we used a single cell layer only). At the bottom of the flow cell, we assumed a constant influx boundary to imitate the effect of the four inflow ports, which were used in the experiment. The flow rate of the influx boundary was set to 0.41 m day$^{-1}$ in order to reproduce the initial head distribution in the observation zone. Results from the preliminary simulation of the entire flow cell were used as a reference (data not shown). The flow rate for the glucose injection port was 0.04 l/day. The time step size was kept constant at 0.001 day in order to fulfill the Courant criterion and to avoid numerical oscillations. Unless stated otherwise, all simulation results presented in this study were calculated using this refined grid.

In the experiment, the longitudinal dispersivity was determined to be $2 \times 10^{-3}$ m, for the transverse dispersivity we assumed a value of $2 \times 10^{-4}$ m. Initial values for hydraulic conductivity and porosity were $2 \times 10^{-3}$ m/s and 0.39, respectively, as determined in the experiment.
4.3. Reactive processes

To simulate the consumption of nitrate and glucose as well as the production of biomass, we assumed that the oxidation of glucose with nitrate, given by

\[ 5C_6H_{12}O_6 + 24NO_3^- + 24H^+ \rightarrow 30CO_2 + 12N_2 + 42H_2O \]  

is catalyzed by bacteria. The total biomass was assumed to consist of two components, bacteria and EPS. As in the experiment bacterial carbon only contributed 5\% of the total organic carbon, we assumed that the growing bacteria also produced EPS at a production rate...
of 20 times the rate of bacterial growth. The parameters used for the simulation of the reactive processes are given in Table 1. For the bacterial carbon density we took the same value as in Thullner et al. (2002a), and for the density of the EPS we took the smallest value for dry mass per wet volume presented in Characklis and Marshall (1990) and assumed a dry mass carbon content of 50%. All other parameters given in Table 1 were fitted to get an optimal reproduction of the measurements such as observed biomass buildup in the flow cell, biomass concentration at the end of the experiment, and nitrate concentration at the outflow. For the latter, results from preliminary simulations were used. With the parameters given in Table 1, simulated nitrate concentration at the outflow of the flow cell was similar to values reported in Thullner et al. (2002a), showing only negligible variations between the different models (data not shown). As observed in the experiment, biomass also accumulated close to the outflow port, where nitrate and glucose were forced to mix. Therefore, a large fraction of the total nitrate consumption occurred at that place. With the refined model only a subdomain of the flow cell was simulated. Therefore, a comparison between measured and simulated concentration at the outflow was not possible.

![Fig. 4. Distribution of biomass volume: (A) measured and (B) simulated in the observation zone for the different hydraulic \( k_i-n \) relations (for the simulation results given in percent of initial porosity).](image-url)
As mentioned, the parameters describing the microbial growth were fitted to reproduce the experimental results. However, the determined parameter values were in a realistic range: $\mu_{\text{max}}$ was similar to values obtained in preliminary batch experiments from the
selected microbial strain with glucose (data not shown) and values for $\mu_{\text{dec}}, K_{\text{nit}}, K_{\text{glu}},$ and $\lambda$ were similar to values used by Clement et al. (1997) for simulating the growth of bacteria on acetate and nitrate in sand columns (after transferring them into the same units). The values from Clement et al. (1997) were also used by Kildsgaard and Engesgaard (2002) for simulating their clogging experiment.

4.4. Bacterial clogging

Parameter values of $a = -1.7, k_{f_{\text{min}}} = 0.01,$ and $b = 1.8$ in Eqs. (7) and (8) were taken from the most homogeneous pore size distribution (standard deviation of $\sigma = 0.33$ for the lognormal pore radii distribution) presented in Thullner et al. (2002b), assuming that the nearly constant diameter glass bead packing used in Thullner et al. (2002a) is best represented by this case. In Thullner et al. (2002b), $n_0$ has been determined for the different growth morphologies, showing much smaller values for the biofilm model (approximately between 0.2 and 0.4) than for the colony model (approximately between 0.7 and 0.9). However, within this study, we allowed $n_0$ to have values different from those reported in Thullner et al. (2002b) and tried to determine if and for what values the models were able to reproduce the experimental observations. In addition, using the same value for $n_0$ in Eqs. (7) and (8) allowed us to investigate whether the simulation results were sensitive against the mathematical expression used to describe the clogging.

5. Results

5.1. Biomass distribution

For all models the general pattern of biomass distribution was similar, showing a V- to U-shaped structure starting close to the glucose injection port (Fig. 4). The biomass concentration showed a gradual increase until approximately day 23 and only small changes between days 23 and 30. Highest peak values for biomass volumes (36–37% of initial porosity filled with biomass) were reached with simulations assuming no clogging or using the Clement model. Using the colony or biofilm model (with a value of $n_0 = 0.75$ in Eqs. (7) and (8), respectively) resulted in smaller peak values for biomass volumes of approximately 24%. This resulted in minimum values for $k_{f_{\text{rel}}}$ of 0.243 (Clement model), 0.0171 (Colony model with $n_0 = 0.75$), and 0.0142 (Biofilm model with $n_0 = 0.75$).

In addition, Fig. 5 shows the biomass distributions at day 30 simulated with the colony and the biofilm model for different values of $n_0$. With one exception, the general pattern was similar in both models for all values of $n_0$. Only for the colony model with $n_0 = 0.9$ the result was slightly different. In that simulation, the distance between the two branches of the biomass distribution pattern slightly exhibited a local minimum a few cm above the glucose injection port. For all other simulations, this distance was constantly increasing between the glucose injection port and the top of the observation zone. Peak values for biomass volumes decreased for increasing $n_0$. For the colony model the highest biomass volumes were 34% of initial porosity for $n_0 = 0.6$ and 12% of initial porosity of $n_0 = 0.9$. 
Fig. 5. Distribution of biomass volume (given in percent of initial porosity) the observation zone at day 30 for the colony and biofilm model using different values of $n_0$. 
Fig. 6. Organic carbon concentration at the end of the experiment for the different $k_f-n$ relations compared to measured data. $x$-axes show distance to left border of flow cell.
Fig. 7. Organic carbon concentration at the end of the experiment for the colony and biofilm model using different values of $n_0$. $x$-axes show distance to left border of flow cell.
For the biofilm model the highest biomass volumes were 33% of initial porosity for $n_0 = 0.6$ and 22% of initial porosity of $n_0 = 0.9$.  

5.2. Organic carbon concentration

To compare simulation results with data from Thullner et al. (2002a), we calculated the average organic carbon (from biomass) concentration at the end of the experiment for the porous medium sampling points shown in Fig. 3, which corresponded to porous media samples analyzed by Thullner et al. (2002a). This was done by transferring the simulated concentrations (expressed as mass carbon per volume) into the units used in the experiment (mass carbon per mass glass bead matrix) using the carbon density of the biomass (input parameter) and the measured bulk density of the glass bead matrix. Results of this procedure are given in Fig. 6 for the different models (colony and biofilm model with $n_0 = 0.75$) together with the experimental data. For transect A, the organic carbon concentration showed a dual peak pattern for all of the models similar to that of the measured data. Highest simulated concentrations for transect A ranged between 28 $\mu$g/g beads for the biofilm model and 39 $\mu$g/g beads for the model without clogging. For transect B, all the simulations showed a dual peak pattern, too, whereas the measured data showed a single peak in the middle of the transect only. Highest simulated organic carbon concentrations for transect B ranged between 14 $\mu$g/g beads for the model without clogging and 21 $\mu$g/g beads for the colony model. When comparing simulation results of the colony and biofilm model using different values for $n_0$ (Fig. 7), the dual peak pattern for transects A and B could be observed for all simulations, with the highest concentrations depending on $n_0$. Using the colony model, the concentration maximum for transect

![Fig. 8. Tracer migration in the observation zone for the different $k_f-n$ relations: (A) measured and (B) simulated. For the simulation results, grey scales show relative tracer concentration ($c/c_0$). Data show tracer distribution 1, 3, and 5 h after start of tracer injection.](image)
Fig. 8 (continued).
Fig. 9. Tracer migration in the observation zone: (A) for the colony model and (B) for the biofilm model using different values of $n_0$. Grey scales show relative tracer concentration ($c/c_0$). Data show tracer distribution 1, 3, and 5 h after start of tracer injection.
Fig. 9 (continued).
A decreased from 35 µg/g beads \((n_0 = 0.6)\) to 24 µg/g beads \((n_0 = 0.9)\), whereas peak concentrations for transect B were highest (22 µg/g beads) for \(n_0 = 0.8\) and decreased to 20 µg/g beads for \(n_0 = 0.6\) and 18 µg/g beads for \(n_0 = 0.9\). Using the biofilm model, the highest organic carbon concentration was 31 µg/g beads for \(n_0 = 0.6\) and decreased to 21 µg/g beads for \(n_0 = 0.9\). For transect B, peak concentration was around 20 µg/g beads for \(n_0\) between 0.6 and 0.8, but for \(n_0 = 0.9\) only 17 µg/g beads was obtained.

### 5.3. Tracer migration

Simulation results for the tracer test on days 28 and 29 are shown in Fig. 8 for the different models together with the experimental observations. Depending on the model used for simulation, the influence of biomass on the tracer migration was different. Assuming no clogging in the model, tracer flow was straight upward with only dispersive mixing increasing the width of the tracer plume. In comparison, the tracer distribution was only slightly different for the Clement model, but a small increase in the width of the tracer plume, starting close to the glucose injection port, was visible. Results from the colony and biofilm model (both with \(n_0 = 0.75\)) showed that the tracer plume initially (1 h after the start of injection) tended to split into two branches when reaching the vicinity of the glucose injection port where the biomass was located. This initial trend disappeared with further migration of the tracer plume, but the width of the tracer plume was clearly increased. When comparing the simulation results for the tracer migration using the colony and the biofilm model with different values for \(n_0\), we observed that the influence of the biomass on the tracer migration was increasing with increasing \(n_0\) for both models (Fig. 9). For values of \(n_0\) between 0.6 and 0.8, the general pattern of a branched tracer plume (1 h after the start of the tracer injection) and the increased width of the tracer plume (3 h and 5 h after start of the injection) could be observed for all simulations, but these effects became more pronounced for higher \(n_0\) (with only small differences between the colony and the biofilm model).

For \(n_0 = 0.9\), the colony and the biofilm model showed different results. For the colony model the tracer plume was split into two branches even 5 h after start of tracer injection. In contrast, for the biofilm model the tracer did not change the general pattern observed for smaller values of \(n_0\).

### 6. Discussion

#### 6.1. Comparison between measured and simulated data

To compare results from different simulations, the experimental data from Thullner et al. (2002a) were divided into three groups: pattern of biomass distribution in the flow cell, measured organic carbon concentration at the porous media sampling points, and the results from the tracer test conducted on days 28 and 29. In contrast to the nitrate concentration at the outflow, which could be reproduced with all of the models (in preliminary simulations), the model that provided the best fit with the measured data varied from group to group.
The measured pattern of biomass distribution (U-shaped pattern starting at the glucose injection point) as well as the kinetics of biomass growth could generally be reproduced with all models, but only the colony model with $n_0 = 0.9$ (Fig. 5) was able to reproduce at least slightly the local minimum of the distance between the two branches of the pattern, as observed in the experiment. Comparing experimental and simulated data for the organic carbon concentration along transects A and B, the best agreement could be achieved assuming no clogging in the flow cell (Fig. 6). With clogging, the simulation results were still close to the measured values for the Clement model as well as for the colony and the biofilm model for $n_0 \leq 0.8$. For larger values of $n_0$, the decrease in organic carbon concentration between the transects A and B, which was observed in the experiment, could not be reproduced satisfactorily by the simulations. Moreover, the measured single peak pattern of organic carbon concentration could not be found in any of the simulations (Fig. 7).

For the tracer migration, agreement between simulations and experiment was increasing with increasing values for $n_0$. The simulated tracer migration for the colony model ($n_0 = 0.8$) and the biofilm model ($n_0 = 0.8$ or $n_0 = 0.9$) reproduced well the measured tracer migration. The colony model with $n_0 = 0.9$ overestimated the change of tracer migration due to bioclogging, whereas the colony and the biofilm model for $n_0 < 0.7$ but also the Clement model were underestimating the change of tracer migration due to biomass. Although the simulated biomass volumes are slightly larger for the Clement model than for the colony and biofilm models, the hydraulic conductivity reduction is less than an order of magnitude for the Clement model but approximately two orders of magnitude for the colony and biofilm model (with $n_0 = 0.75$), which explains the differences in the simulated tracer migration. Of course, the biomass had no influence on the tracer migration in the model without bioclogging.

Although for the simulation results presented here we did not modify other parameters than $n_0$, it should be pointed out that in additional simulation a modification of parameters describing the reactive transport did not lead to a better agreement between measured and simulated data. Especially, the unsatisfactorily reproduction of the biomass distribution and the organic carbon concentration, respectively, for transect B remained. Data given in Thullner et al. (2002a) showed that for transect B the biomass composition was changing along the transect. Bacterial numbers exhibited a dual peak pattern as simulated by the models, whereas data for EPS, and thus the total biomass showed only a single peak in the middle of the transect. In our simulations, we did not assume changes in the biomass composition, which may be a reason for the differences between simulated and measured data for transect B. Other potential reasons for these differences were assuming the flow field to be strictly two-dimensional, not considering any three-dimensional effects. Moreover, the operator splitting technique used to solve flow, transport, and reactive processes in a consecutive order with updating the clogging-related parameters only once per time step may be an additional an source for simulation errors. However, the TBC model of Schäfer et al. (1998a), which is the underlying model used for this study, was used successfully for reactive transport simulations (without clogging) on various scales (e.g., Schäfer et al., 1998b; Thullner and Schäfer, 1999). In addition, comparing the used time step size (0.001 day) with the maximum growth rate for the bacteria (1 day$^{-1}$) indicates that the changes in biomass concentration per time step were very small. Thus, all subsequent changes in the clogging parameters were small, too, even when considering
the involved nonlinear relations. This indicates that the used numerical scheme is not likely to be the reason for any discrepancies between measured and simulated results. The fact that there were no measured values for the transversal dispersivity available forced us to estimate the latter based on the measured value for the longitudinal dispersivity. As the numerical dispersion observed for the preliminary simulation using a coarser spatial discretization did not allow to simulate the biomass buildup satisfactorily, it must be concluded that the used dispersivities were not selected too low. However, evaluating the effect of smaller dispersivity values on the simulation results (i.e., on the biomass distribution in transect B) would have required an even finer spatial discretization than used to obtain the presented simulation results. As this would also have led to smaller time steps, the computational requirements were to high to perform such an analysis. For the same reasons, it was not appropriate to simulate the entire flow cell using the refined grid. This made it necessary to approximate some of the boundary conditions used for simulating the presented results. I.e., we had to assume nonpoint sources for the background flow and we also had to ignore lateral flow across the vertical boundaries of the refined grid. However, for the refined grid the flow rate of the glucose injection port was only 6% of the background flow rate (compared to 2% for the entire flow cell) and there were no indications that these approximations had any negative impact on the accuracy of the simulation results.

6.2. Evaluation of clogging models

6.2.1. Comparison of different clogging models

To decide which model allowed the best reproduction of all data groups measured by Thullner et al. (2002a), we had to find a compromise that reproduced all data groups satisfactorily. Considering that the tracer migration was definitely changed by the biomass in the experiment, the models that did not reproduce this effect at least partly were assumed to be inappropriate for simulating the flow cell experiment. The colony and the biofilm models allowed the best reproduction of the tracer data, especially for $n_0 > 0.7$, but for values of $n_0 \geq 0.8$ the simulated organic carbon concentration for transects A and B did not fit the measured data in an acceptable way. Thus, the best overall fit could be achieved with the colony and the biofilm model with $n_0 = 0.75$.

Although the simulation results from the colony and the biofilm model were very similar for $n_0 < 0.9$, values of $n_0$, for which an appropriate reproduction of the experimental data could be achieved, are closer to values used in Thullner et al. (2002b) for describing the influence of microbial colonies in pore networks. For the colony model the best fit value of $n_0 = 0.75$ agrees well with the value of $n_0 = 0.7$, which was the result for the colony model for the most homogeneous pore networks used in the study of Thullner et al. (2002b). In contrast, for the biofilm model the best fit parameter was also $n_0 = 0.75$, but this is much higher than the values determined for the biofilm model in the study of Thullner et al. (2002b). In the latter study, results for $n_0$ vary between 0.2 and 0.4 depending on the heterogeneity of the pore networks. As the biofilm model was clearly underpredicting the observed clogging effects already for $n_0 = 0.6$, a further reduction of this parameter to values determined by the pore network simulations would obviously not allow reproduce the clogging effects observed in the experiment of Thullner et al. (2002a).
This indicates that using the biofilm model with parameter values proposed by the network simulations of Thullner et al. (2002b) does not properly explain the bioclogging observed in this experiment.

This suggested that the assumption of biomass growing in colonies as described in Thullner et al. (2002b) was better suited to describe the flow cell experiment. Moreover, results of this study agree with the observations of Thullner et al. (2002b), who reported a better agreement between the colony model and experimental data (taken from the literature) in case of porous media having a grain diameter of less than 1 mm. Nevertheless, a larger number of experimental data sets would be desirable to further clarify this question and to decide whether this is a general finding.

Our results confirm the assumption of Vandevivere et al. (1995) in that models, which account for pore connectivity in more than one dimension and, especially, which assume biomass growth not only in a biofilm but in form of colonies, may allow better predictions than models as those of Clement et al. (1996) and Taylor et al. (1990). We showed that the colony model, which was derived from pore network simulations, was able to reproduce measured clogging effects, whereas the Clement model was underestimating the bioclogging as well as the biofilm model when using the parameters presented in Thullner et al. (2002b).

6.2.2. Sensitivity of parameters

Results from this study showed that an experimental data set, which included measurements of biomass distribution and the flow field but did not include measurement of hydraulic conductivity changes, could be used to evaluate a model including bioclogging. As the measurement of local hydraulic conductivities may be impossible in multi-dimensional systems this finding is of importance. In contrast, this study also showed the importance of measuring a variety of different data in order to make an accurate evaluation of a clogging model, because each of the data groups was best reproduced with a different model and the combination of all data groups was needed to decide, which model was the best compromise. This may also explain the fact that Kildsgaard and Engesgaard (2001) were able to simulate the clogging experiment of Kildsgaard and Engesgaard (2002) using the Clement model. Although they quantified the tracer distribution in their experiment, no biomass measurements were available for their experiment, and thus, the biomass could be used as a fit parameter in their simulations.

For transforming the mass of EPS into volume of EPS we had to make an assumption on the density of EPS. As reviewed by Characklis and Marshall (1990), values for this density (expressed in dry weight per wet volume) vary between 5 and 130 kg/m³. For the density of EPS in this study we decided to use the smallest value of 5 kg/m³ (which corresponds to 2.5 kg C/m³), giving the EPS and therefore the total biomass a maximum of volume. In case that the density of the EPS was higher in the flow cell experiment than assumed for the simulations, the total volume of biomass would have been smaller than simulated in this study. To explain the observed clogging effects in such a case would be possible by adjusting $n_0$ towards higher values in the colony or biofilm model, leading to even more unrealistic values for the biofilm model. The Clement model, which already underestimated the bioclogging effects for small EPS densities, would by far not be able to explain the bioclogging effects for high EPS densities, again indicating that the colony
model is more appropriate than the biofilm model and, especially, the Clement model to describe bioclogging in porous media. As the contribution of bacterial volume to total biomass volume was negligible, simulation results were not sensitive to the density of bacteria.

We did not explicitly investigate the sensitivity of the simulation results towards variations of parameters other than \( n_0 \) in Eqs. (7) and (8). Nevertheless, small differences between the colony and biofilm model for a given \( n_0 \) indicated that simulation results were not sensitive to the expression of the \( k_f \) decrease for porosity reductions between \( n = 1 \) and \( n = n_0 \). This indicates that the sensitivity of the simulation results towards the parameters mentioned above must have been small, too, as most of these parameters only modify the \( k_f \) decrease between \( n = 1 \) and \( n = n_0 \), but they do not alter the position of the minimal \( k_f \). Practically, this also means that knowing the appropriate value for \( n_0 \) is much more relevant for the description of bioclogging than finding the best mathematical expression for describing the decrease of \( k_{f_{rel}} \) between \( n_{rel} = 1 \) and \( n_{rel} = n_0 \). As already pointed out above, a more detailed investigation of the sensitivity of the simulation results towards changes in (transversal) dispersivity could not be performed, although we can exclude higher dispersivities leading to a better reproduction of the measured data. As the mixing of nitrate and glucose was controlled by dispersive processes, it must be assumed that the biomass growth in this mixing zone, and thus the clogging, were sensitive to the used dispersivity parameters. However, this would have an influence on the biomass buildup in general and not on the clogging effect caused by a given amount of biomass.

Finally, the simulation results showed that two parameters were important for the description of bioclogging. The first one is the density of EPS or more general the density of the biomass. As the volume of a given amount of biomass directly depended on its density, simulated porosity reductions were highly sensitive to the biomass density. The second parameter was \( n_0 \) in Eqs. (7) and (8). This parameter contains the information of how much biovolume was needed to clog the porous medium. These results suggest that experimental determination of biomass densities and how much biovolume is present in a clogged porous medium would strongly reduce the uncertainty of the numerical simulations describing the reduction of hydraulic conductivity, especially because the sensitivity of the simulations results was low to other parameters.

### 7. Conclusion

Results from this study showed that it was generally possible to simulate the experimental results from *Thullner et al. (2002a)* with a numerical model. A detailed comparison between the experimental data and the simulation results showed that best reproduction could be achieved with a model assuming microbial growth in colonies, while a biofilm model was able to simulate the experimental data for unrealistic parameter values only (both models derived from pore network simulations). The model published by *Clement et al. (1996)* was strongly underestimating the clogging effects observed in the experiment. This finally suggests that it is necessary to assume biomass growth in discontinuous colonies rather than a homogeneous biofilm, and to include multi-dimen-
sionality effects already on the pore scale in order to get an appropriate description of bioclogging in porous media.

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