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A mathematical model and analytical solution for the fixation of bacteria in Biogrout

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Abstract

Biogrout is a new method for soil reinforcement, which is based on microbial induced carbonate precipitation. Bacteria and reagents are flushed through the soil, resulting in calcium carbonate precipitation and consequent soil reinforcement. Bacteria are essential in the Biogrout process since they catalyse the reaction. Hence, to control the process, it is essential to know where the bacteria are situated. The bacteria are possibly in suspension but can also be adsorbed or fixated on the matrix of the porous structure. In this paper, a model is derived for the placement of bacteria. The model contains three phases of bacteria: bacteria in suspension, adsorbed bacteria and fixed bacteria. An analytical solution is derived for instantaneous reactions between these three phases. The analytical solution is compared to numerical simulations for finite reaction rates. For the numerical simulations the Standard Galerkin Finite Element Method is used.

1 INTRODUCTION

1.1 Soils on demand

The mechanical properties of soil (cohesion, friction, stiffness and permeability) are essential for engineering constructions. Nowadays, several techniques are being developed, which change the soil properties on demand. This is done by stimulating biochemical processes in situ [DeJong et al. 2010, Ivanov et al. 2008, Van Meurs et al. 2006]. Biogrout is one of these techniques and it is based on microbial-induced carbonate precipitation (MICP).

Several researchers have shown that MICP can be used to improve the mechanical properties of porous materials, [Bang et al. 2001, Bachmeier et al. 2002, Nemati et al. 2003, DeJong et al. 2006, Whiffin et al. 2007].

The Biogrout process has been studied for a couple of years, [Whiffin 2004, Whiffin et al. 2007, Van Paassen 2009, Van Paassen et al. 2009, Van Paassen et al. Oct. 2009, Van Paassen et al. 2010, Harkes et al. 2010, Van Wijngaarden et al. 2010, Van Wijngaarden et al. 2011], in which the last two mentioned studies are computational.

1.2 What is Biogrout?

Biogrout is based on microbial-induced carbonate precipitation. In this article, we focus on the *Sporosarcina pasteurii* bacteria. These bacteria contain the enzyme *urease*, that provides the hydrolysis of urea ($CO(NH_2)_2$). The reaction equation for the hydrolysis of urea is given by (see [Whiffin et al. 2007])

$$CO(NH_2)_2(aq) + 2H_2O(l) \overset{\text{bacteria}}{\longrightarrow} 2NH_4^+(aq) + CO_3^{2-}(aq). \tag{1}$$

The products of this reaction are carbonate (CO_3^{2-}) and the by-product ammonium (NH_4^+). In the presence of calcium ions (Ca^{2+}) and if the solution is oversaturated, the calcium and carbonate

ions precipitate as calcium carbonate ($CaCO_3$). This happens in several steps, depending on the pH. The overall precipitation reaction is given by (see also [Whiffin et al. 2007])

$$Ca^{2+}(aq) + CO_3^{2-}(aq) \rightarrow CaCO_3(s). \tag{2}$$

Combining the hydrolysis reaction equation (1) and the precipitation reaction equation (2) gives the overall Biogrout reaction equation:

$$CO(NH_2)_2(aq) + Ca^{2+}(aq) + 2H_2O(l) \rightarrow 2NH_4^+(aq) + CaCO_3(s). \tag{3}$$

When applying Biogrout, first of all the bacteria are cultivated. Subsequently, the bacteria are injected into the subsoil and transported by water flow to the location where strengthening is required. The bacterial suspension is directly followed by a fixation fluid, which is a solution with high salinity. As a result of the retardation of the bacteria, the fixation fluid will overtake the weakly adsorbed bacteria and strongly fix them to the soil particles [Harkes et al. 2010]. This will result in a rather homogeneous distribution of bacteria. After the placement of the bacteria, the urea and calcium chloride ($CaCl_2$) solution is supplied [Whiffin et al. 2007]. Due to the calcium chloride, this solution has an high salinity and will therefore also act as a fixation fluid. The bacteria provide the hydrolysis of urea and the calcium and carbonate ions precipitate as calcium carbonate. The solid calcium carbonate forms bridges between the sand grains, in this way, the strength and stiffness of the soil increase. The by-product ammonium chloride (NH_4Cl) needs to be removed. The bacteria and reactants are not injected at the same time to prevent clogging and crystal accumulation around the injection point(s). The procedure, in which the bacteria are first injected and only then followed by the reactants, also results in a more homogeneous distribution of calcium carbonate.

1.3 Transport of bacteria

Since the bacteria provide the hydrolysis of urea, they are crucial in the Biogrout process. The reaction rate increases with an increasing bacterial concentration and urease activity. Further, if no bacterium is present, no carbonate is formed and consequently no calcium carbonate appears. Therefore, it is essential to know where the bacteria are situated.

When modelling bacterial transport, it is not sufficient to consider advection and dispersion only. Adsorption and desorption are important phenomena as well, but also the pore size of the matrix, the size of the microorganisms, filtration and elimination ([Matthess et al. 1981, Matthess et al. 1988, Fontes et al. 1991, Foppen et al. 2006]), ionic strength of the ground water ([Fontes et al. 1991, Foppen et al. 2006]), systematic (chemotaxis) and random (tumbling) motion of bacteria ([Yavuz Corapcioglu et al. 1984]), residence time ([Johnson et al. 1995]), decay and growth ([Yavuz Corapcioglu et al. 1984, Foppen et al. 2006]) effect the (rate of) transport of microorganisms.

[Hornberger et al. 1992, Johnson et al. 1995, Tan et al. 1994] provide models that consider several of these phenomena and compare the model results with experimental results.

1.4 Applications of Biogrout

The Biogrout process can be used in a wide variety of situations, in which it is desirable to change the properties of the subsoil. We mention the following examples

- prevention of liquefaction [Ruyt et al. 2009, DeJong et al. 2010];
- bore hole stabilization [Star et al. 2011];

- slope stabilization [DeJong et al. 2010];
- stabilization of railroad tracks [Van Paassen 2009];
- reinforcement of calcarenite room and pillar mines [Van Paassen et al. 2008].

This motivates the value to investigate Biogrout.

1.5 Scope of this article

In [Van Wijngaarden et al. 2010] and [Van Wijngaarden et al. 2011], the study is focussed on modelling the transport of the reactants, assuming a homogeneous distribution of bacteria. The present study is devoted to the transport of bacteria. In Section 2 a model is derived for the placement of bacteria. Further, initial and boundary conditions are given. In Section 3 the analytical solution for the (simplified) model equations derived in Section 2 for the instantaneous case is presented. In Section 3 the Numerical Methods to solve the model equations are described. In Section 4, the results are displayed and a comparison is made between the analytical solution and the numerical solutions. In the last section, some conclusions and a discussion can be found.

2 Mathematical model

2.1 Derivation of the model equations

First we present the general equation for the transport of bacteria in a fully saturated porous medium, as in for example [Tan et al. 1994]:

$$\frac{\partial(\theta C)}{\partial t} + \frac{\partial\left(\theta C^{adsorbed}\right)}{\partial t} = \nabla\cdot(D_{bac}\theta\nabla C) - \nabla\cdot(\mathbf{q}C)\,. \tag{4}$$

In this equation, θ is the porosity, C is the bacterial concentration in suspension, $C^{adsorbed}$ is the imaginary concentration that would result if the attached bacteria were to be resuspended in a solution volume equivalent to that of the surrounding water. Note that in literature, $C^{adsorbed}$ is frequently expressed in units of milligrams per kilogram, see for example [Zheng and Bennett 1995]. Here, $C^{adsorbed}$ has the same unit as C. Further, D_{bac} is the dispersion coefficient of bacteria in suspension and $\mathbf{q}$ is the Darcy velocity, which relates to the pore water flow velocity $\mathbf{v}$ as $\mathbf{q} = \mathbf{v}\theta$. The terms at the left-hand side are the accumulation terms for the suspended and adsorbed bacteria. The first term at the right-hand side accounts for dispersion and the last term is the advection term. Since bacterial growth and decay are processes with a large time scale we neglect both last mentioned phenomena. Further, we assume bacterial movement to be determined by flow only, which means that their systematic movement is neglected.

In the case of an equilibrium-controlled adsorption, $C^{adsorbed}$ tends to the equilibrium $\varphi(C)$, where φ is an adsorption isotherm, which depends on the concentration of bacterial cells in suspension (C) and might also depend on properties of the microorganisms and the porous medium and the pH. To be able to calculate C and $C^{adsorbed}$ separately, equation (4) is split into two equations:

$$\frac{\partial(\theta C)}{\partial t} = \nabla\cdot(D_{bac}\theta\nabla C) - \nabla\cdot(\mathbf{q}C) - \theta r_{ads} + \theta r_{des}, \tag{5}$$

$$\frac{\partial\left(\theta C^{adsorbed}\right)}{\partial t} = \theta r_{ads} - \theta r_{des}, \tag{6}$$

where

$$r_{ads} = k_{ads}\left(\varphi(C) - C^{adsorbed}\right)_+, \tag{7}$$

$$r_{des} = k_{des}\left(C^{adsorbed} - \varphi(C)\right)_+. \tag{8}$$

The reaction r_{ads} is the adsorption reaction, r_{des} is the desorption reaction, k_{ads} and k_{des} are respectively the adsorption and desorption rate constant. The notation $(.)_+$ considers the positive part of an expression and has been defined as $(.)_+ := \max(0, .)$. Equations (7) and (8) account for the difference in desorption and adsorption rate. If there are no bacteria in suspension, no bacteria can adsorb, therefore $\varphi(0) = 0$. The more bacteria in suspension, the more bacteria will adsorb, so $\varphi'(C) > 0$ and hence $\varphi(C) > 0$. It is also assumed that $\varphi''(C) < 0$ and $\lim_{C \to \infty} \varphi'(C) = 0$, which implies that the adsorption rate decreases as adsorption proceeds. This is a logical result from the fact that a higher number of adsorbed bacteria gives a lower number of free adsorption sites.

In the Biogrout process, the bacterial suspension is directly followed by a fixation fluid, which is a solution with high salinity. This fixation fluid will overtake the weakly adsorbed bacteria and strongly fix them onto the solid matrix. In order to model this, $C^{adsorbed}$ is split up into a temporarily adsorbed part $\overline{C}$ and a permanently adsorbed, or fixed, part S:

$$C^{adsorbed} = \overline{C} + S. \tag{9}$$

In the case of an equilibrium, the concentration of temporarily adsorbed bacteria $\overline{C}$ is given by $\overline{C} = (1 - \beta)\varphi(C)$ and, since S is the concentration of *permanently* adsorbed bacteria, which can not decrease, the following equilibrium holds: $S(\mathbf{x}, t) = \max_{0 \leq \bar{t} \leq t} \{\beta\varphi(C(\mathbf{x}, \bar{t}))\}$. From this equation follows that, for a constant C, $S = \beta\varphi(C)$. The fraction β ranges between 0 and 1, where the value depends on the concentration of the fixation fluid and may also depend on, for example, properties of the microorganisms, the pH and the porous medium.

Substituting relation (9) into the equations (6), (7) and (8), gives the following equations for the adsorbed bacteria:

$$\frac{\partial\left(\theta\left(\overline{C} + S\right)\right)}{\partial t} = \theta r_{ads} - \theta r_{des}, \tag{10}$$

$$r_{ads} = k_{ads}\left(\varphi(C) - \left(\overline{C} + S\right)\right)_+, \tag{11}$$

$$r_{des} = k_{des}\left(\overline{C} + S - \varphi(C)\right)_+ \tag{12}$$

$$= k_{des}\left(\left(\overline{C} - (1 - \beta)\varphi(C)\right) + \left(S - \beta\varphi(C)\right)\right)_+. \tag{13}$$

Next, we show that equation (13) needs to be adjusted. According to this equation, both the difference between $\overline{C}$ and its equilibrium and the difference between S and its equilibrium, are driving forces for desorption. Consider now the situation $\overline{C} + S > \varphi(C)$, $\overline{C} < (1 - \beta)\varphi(C)$ and $S > \beta\varphi(C)$, which can happen when C (and hence $\beta\varphi(C)$) is decreasing and when the adsorption rate r_{ads} is not so high. Compared to the equilibrium $\beta\varphi(C)$, too many bacteria are adsorbed, and according to equation (13) there is a driving force for desorption. Concentration S is the concentration of permanently adsorbed bacteria and these bacteria will not desorb again. This implies that the concentration temporarily adsorbed bacteria $\overline{C}$ will decrease. However, this concentration is already lower than its equilibrium $(1 - \beta)\varphi(C)$. This means that the difference between S and its equilibrium $\beta\varphi(C)$ will result in extra desorption and this is not allowed. Therefore, the difference between S and its equilibrium should only be taken into account if $S < \beta\varphi(C)$. In that case, it will diminish desorption. Hence, equation (13) is replaced by

$$r_{des} = k_{des}\left(\left(\overline{C} - (1 - \beta)\varphi(C)\right) + \left(S - \beta\varphi(C)\right)_-\right)_+, \tag{14}$$

in which the notation $(.)_-$ considers the negative part of an expression and is defined by $(.)_- := \min(0, .)$.

For the concentration of permanently adsorbed bacteria S, the following equation is used:

$$\frac{\partial\left(\theta S\right)}{\partial t} = \theta r_{fix}, \tag{15}$$

in which the fixation rate r_{fix} is given by

$$r_{fix} = k_{fix}\overline{C}\left(\beta\varphi(C) - S\right)_+. \tag{16}$$

The constant k_{fix} is the fixation rate constant. The driving force for fixation is the difference between the concentration of fixated bacteria S and its equilibrium $\beta\varphi(C)$, which is accounted for by the term $(\beta\varphi(C) - S)_+$. Only the positive part of this expression is taken into account, since S is the concentration *permanent* adsorbed bacteria, which can not decrease. If there are no adsorbed bacteria, they can not be fixated. If there are many adsorbed bacteria it is likely that fixation proceeds faster than in the case that there are only a few adsorbed bacteria. That is the reason why the fixation rate also contains a multiplication by $\overline{C}$.

From equations (10) and (15), the following differential equation is found for the concentration temporarily adsorbed bacteria:

$$\frac{\partial\left(\theta\overline{C}\right)}{\partial t} = \theta r_{ads} - \theta r_{des} - \theta r_{fix}. \tag{17}$$

For the concentration of the fixation fluid c^{fix}, the following differential equation is used:

$$\frac{\partial(\theta c^{fix})}{\partial t} = \nabla\cdot(D_{fix}\theta\nabla c^{fix}) - \nabla\cdot(\mathbf{q}c^{fix}), \tag{18}$$

in which D_{fix} is the dispersion coefficient of the fixation fluid.

To summarize, we solve the following system of equations for the transport of bacteria in a saturated porous medium, in combination with a fixation fluid.

$$\begin{aligned}
\frac{\partial\left(\theta C\right)}{\partial t} &= \nabla\cdot(D_{bac}\theta\nabla C) - \nabla\cdot(\mathbf{q}C) - \theta r_{ads} + \theta r_{des}, && (19)\\
\frac{\partial\left(\theta\overline{C}\right)}{\partial t} &= \theta r_{ads} - \theta r_{des} - \theta r_{fix}, && (20)\\
\frac{\partial\left(\theta S\right)}{\partial t} &= \theta r_{fix}, && (21)\\
r_{ads} &= k_{ads}\left(\varphi(C) - \left(\overline{C} + S\right)\right)_+, && (22)\\
r_{des} &= k_{des}\left(\left(\overline{C} - (1-\beta)\varphi(C)\right) + \left(S - \beta\varphi(C)\right)_-\right)_+, && (23)\\
r_{fix} &= k_{fix}\overline{C}\left(\beta\varphi(C) - S\right)_+, && (24)\\
\frac{\partial(\theta c^{fix})}{\partial t} &= \nabla\cdot(D_{fix}\theta\nabla c^{fix}) - \nabla\cdot(\mathbf{q}c^{fix}). && (25)
\end{aligned}$$

2.2 Initial conditions and boundary conditions

For the concentration of suspended, adsorbed and fixed bacteria and for the concentration of the fixation fluid, the following initial conditions are chosen:

$$C(x,0) = \overline{C}(x,0) = S(x,0) = c^{fix}(x,0) = 0. \tag{26}$$

At time $t = T_0$ injecting bacteria is stopped and from then on fixation fluid is injected. This results in the following boundary condition for the concentration of suspended bacteria:

$$C(0,t) = \begin{cases} 1 & \text{for } 0 < t < T_0, \\ 0 & \text{for } t > T_0, \end{cases} \tag{27}$$

and for the concentration of fixation fluid:

$$c^{fix}(0,t) = \begin{cases} 0 & \text{for } 0 < t < T_0, \\ 1 & \text{for } t > T_0. \end{cases} \tag{28}$$

3 Analytical Solution and Numerical Methods

In this section, the analytical solution for the simplified version of system (19)-(25) is derived. A case study is presented for one particular adsorption isotherm. The model equations have also been solved numerically. The numerical strategy is described at the end of this section. The following simplifications have been made:

- Restriction to one dimension;
- Dispersion and diffusion are neglected: $D_{bac} = D_{fix} = 0\ m^2/h$;
- The pore water velocity is equal to 1: $v = 1\ m/h$;
- The porosity θ is constant and can be taken out of the differential equations by division;
- For β the following has been chosen: $\beta = \beta_0 c^{fix}$, in which β_0 is a constant;
- The adsorption isotherm only depends on the concentration suspended bacteria: $\varphi = \varphi(C)$.

Note that using different values for θ and $\mathbf{q}$ (and $\mathbf{v}$) is trivial with respect to scaling.
These simplifications result in the following system of equations:

$$\frac{\partial C}{\partial t} = -\frac{\partial C}{\partial x} - r_{ads} + r_{des}, \tag{29}$$

$$\frac{\partial \overline{C}}{\partial t} = r_{ads} - r_{des} - r_{fix}, \tag{30}$$

$$\frac{\partial S}{\partial t} = r_{fix}, \tag{31}$$

$$r_{ads} = k_{ads}\left(\varphi(C) - \left(\overline{C} + S\right)\right)_+, \tag{32}$$

$$r_{des} = k_{des}\left(\left(\overline{C} - (1-\beta)\varphi(C)\right) + \left(S - \beta\varphi(C)\right)_-\right)_+, \tag{33}$$

$$r_{fix} = k_{fix}\overline{C}\left(\beta\varphi(C) - S\right)_+, \tag{34}$$

$$\frac{\partial c^{fix}}{\partial t} = -\frac{\partial c^{fix}}{\partial x}. \tag{35}$$

The initial and boundary conditions are given in Section 2.2.

3.1 Analytical solution

In this subsection, we describe the analytical solution for the various components: the fixation fluid and the bacteria.

3.1.1 Fixation fluid

Before deriving the analytical solution for the bacteria, first the solution for the fixation fluid is derived. A solution of (35) is $c^{fix}(x-t)$. Combining this with the initial and boundary condition gives the following solution for the fixation fluid:

$$c^{fix}(x,t) = H((t - T_0) - x), \tag{36}$$

where $H : \mathbb{R} \to \{0, 1\}$ represents the Heaviside function, given by

$$H(y) = \begin{cases} 1 & \text{for } y > 0, \\ 0 & \text{for } y < 0. \end{cases} \tag{37}$$

3.1.2 Bacteria

For the derivation of the analytical solution for the concentration of suspended, adsorbed and fixed bacteria, an instantaneous equilibrium has been assumed: $k_{ads} \to \infty$, $k_{des} \to \infty$ and $k_{fix} \to \infty$. As a result, $\overline{C}$ and S can be found directly as a function of C:

$$\overline{C} = (1 - \beta_0 c^{fix})\varphi(C), \tag{38}$$

$$S = \max_{0 \le \bar{t} \le t} \left\{ \beta_0 c^{fix} \varphi(C) \right\}. \tag{39}$$

The total bacterial concentration Ψ is defined as $\Psi := C + \overline{C} + S$. Adding the differential equations for C, $\overline{C}$ and S ((29), (30) and (31)), gives the following differential equation for Ψ:

$$\frac{\partial \Psi(C)}{\partial t} = -\frac{\partial C}{\partial x}. \tag{40}$$

It is assumed that C is piecewise continuously differentiable in t and x and that φ and Ψ are continuous functions in C.

Along characteristics, we have

$$0 = \frac{d}{dt} C(t, x(t)) = C_t + C_x x'(t) \tag{41}$$

and hence

$$x'(t) = \frac{1}{\Psi'(C)}, \tag{42}$$

in which $\Psi'(C)$ is given by

$$\Psi'(C) = \begin{cases} 1 + \varphi'(C) & \text{if } c^{fix}(x,t)\varphi(C(x,t)) > \max\limits_{0 \le \bar{t} < t} \left\{ 0, c^{fix}(x,\bar{t})\varphi(C(x,\bar{t})) \right\}; \\ 1 + (1 - \beta_0 c^{fix})\varphi'(C) & \text{else.} \end{cases} \tag{43}$$

This results in a sketch of the (x-t)-diagram as displayed in Figure 1.

The velocity of the characteristics originating for the x-axis, where $C = c^{fix} = 0$, is calculated with (42) and (43):

$$x'(t) = \frac{1}{\Psi'(0)} = \frac{1}{1 + \varphi'(0)}. \tag{44}$$

For the characteristics, originating from the t-axis, we distinguish between $0 < t < T_0$ and $t > T_0$. For $t > T_0$, since $C = 0$ and $c^{fix} = 1$ at the inflow boundary, the velocity of the characteristics is given by:

$$x'(t) = \frac{1}{\Psi'(0)} = \frac{1}{1 + (1 - \beta_0)\varphi'(0)}. \tag{45}$$

For $0 < t < T_0$, since $C = 1$ and $c^{fix} = 0$ at the inflow boundary, the following expression is found for the velocity of the characteristics, originating from the t-axis:

$$x'(t) = \frac{1}{\Psi'(1)} = \frac{1}{1 + (1 - \beta_0 c^{fix})\varphi'(1)}. \tag{46}$$

Note that the velocity changes after intersection with the characteristic of the fixation fluid, that starts in $(0, T_0)$. Below this characteristic, we have $c^{fix} = 0$ and therefore $x'(t) = \frac{1}{1+\varphi'(1)}$. Above this characteristic, we have $c^{fix} = 1$ and $x'(t) = \frac{1}{1+(1-\beta_0)\varphi'(1)}$.

The characteristics from $(0, T_0)$ have a lower velocity than the characteristics originating from the x-axis. This is the result of the retardation effect due to adsorption and it results in a shock. The shock position is denoted by $s(t)$. Let $t = T_1$ be the time at which the front of the fixation fluid reaches the front of the pulse with bacteria. For $0 < t < T_1$, the shock speed $s'(t)$ is determined by the Rankine-Hugoniot condition (in which [.] means the jump over the quantity):

$$s'(t) = \frac{[C]}{[\Psi(C)]} = \frac{[C]}{[C + (1 - \beta_0 c^{fix})\varphi(C)]} = \frac{1}{1 + \varphi(1)} < 1. \tag{47}$$

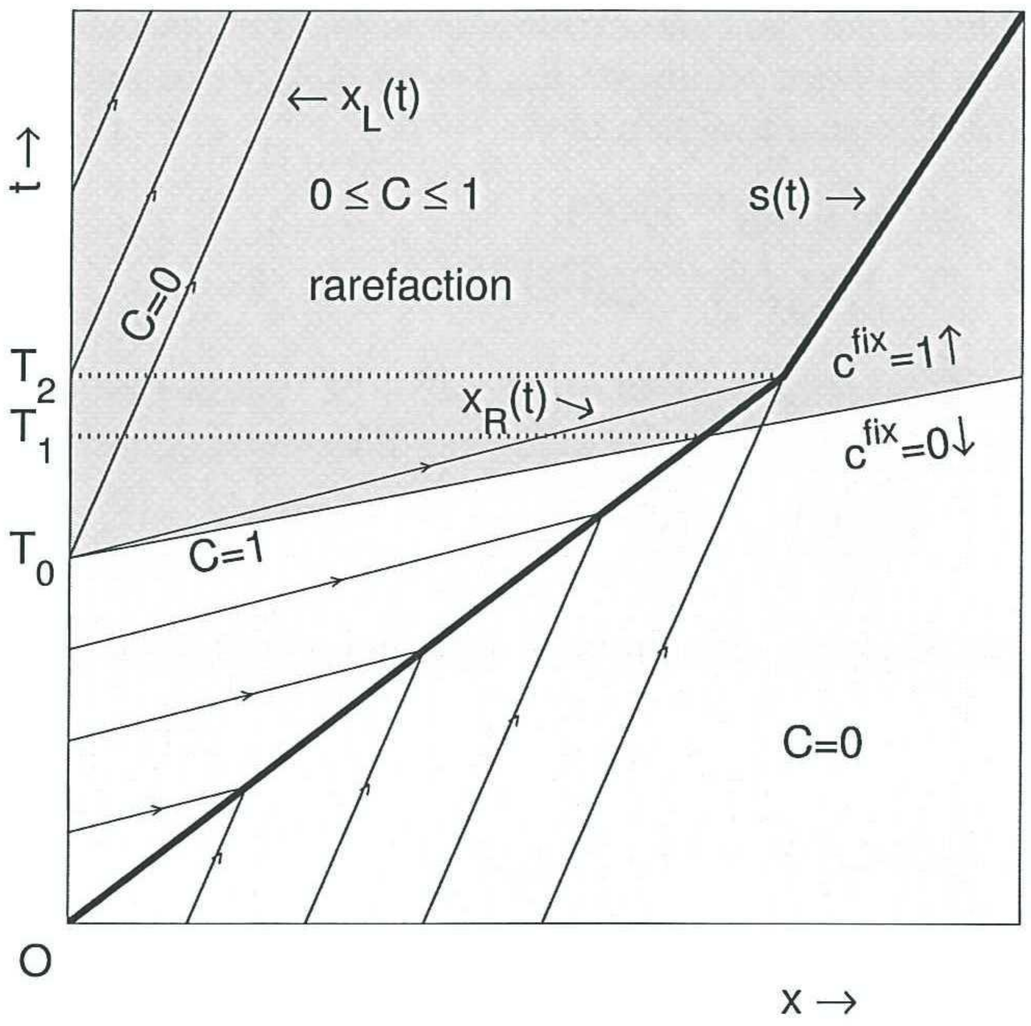

Figure 1: Sketch of the (x-t)-diagram.

Since $s(0) = 0$, for $0 < t < T_1$ the shock position is given by

$$s(t) = \frac{t}{1+\varphi(1)}. \tag{48}$$

From the intersection of the shock position with the position of the fixation fluid front, T_1 can be found: $\frac{T_1}{1+\varphi(1)} = T_1 - T_0$. Hence,

$$T_1 = \frac{1+\varphi(1)}{\varphi(1)} T_0. \tag{49}$$

Since $\varphi(1)$ is positive, we have that $T_1 > T_0$.

Let $t = T_2$ be the time at which the shock speed changes. This change is the result of the decrease of $C(s(t), t)$. For $T_1 < t < T_2$, the shock speed is given by

$$s'(t) = \frac{[C]}{[\Psi(C)]} = \frac{[C]}{[C + (1-\beta_0 c^{fix})\varphi(C) + \max\limits_{0\le\bar{t}\le t}\{\beta_0 c^{fix}\varphi(C)\}} = \frac{1}{1+\varphi(1)}. \tag{50}$$

For $T_1 < t < T_2$, the same shock speed has been found as for $0 < t < T_1$, as can be seen from equations (47) and (50). While deriving the model for the placement of bacteria, this turned out to be very important, since a change in the shock speed at $t = T_1$ turned out to result in a violation of the conservation of mass requirement.

For $0 < t < T_2$ the shock position is given by

$$s(t) = \frac{t}{1+\varphi(1)}. \tag{51}$$

At $t = T_0$, the boundary condition changes. This gives a rarefaction wave, as can be seen from the *(x-t)*-diagram. To this extent, we use the Ansatz $C(t, x) = \tilde{C}(\eta)$, $\eta = \frac{x}{t-T_0}$. This gives the

following derivatives:

$$\begin{cases} C_t &= -\frac{\eta}{t-T_0}\tilde{C}'(\eta), \\ C_x &= \frac{1}{t-T_0}\tilde{C}'(\eta), \\ (\Psi(C))_t &= -\Psi'(\tilde{C})\tilde{C}'(\eta)\frac{\eta}{t-T_0}. \end{cases} \tag{52}$$

Substituting (52) in (40) gives

$$\left(-\Psi'(\tilde{C})\eta + 1\right)\tilde{C}'(\eta) = 0. \tag{53}$$

This equation admits two states:

$$\begin{cases} 1) & \tilde{C}'(\eta) &= 0 & \text{(constant state)}, \\ 2) & \eta &= \frac{1}{\Psi'(\tilde{C})} & \text{(variable state)}. \end{cases} \tag{54}$$

The variable state implies that

$$\Psi'(\tilde{C}) = \frac{1}{\eta}. \tag{55}$$

Since $\Psi'(\tilde{C}) > 0$ and Ψ is a continuous function in C, this equation can be solved ($\Psi'(\tilde{C})$ is invertible). The solution is $C = (\Psi')^{-1}\left(\frac{1}{\eta}\right)$. The constant states are located at

$$\frac{x_L(t)}{t-T_0} = \eta_L = \frac{1}{\Psi'(0)} < \frac{1}{\Psi'(1)} = \eta_R = \frac{x_R(t)}{t-T_0}. \tag{56}$$

Time T_2 can be found from the intersection point of $s(t)$ and $x_R(t)$: $\frac{T_2}{1+\varphi(1)} = \frac{T_2-T_0}{\Psi'(1)}$. Solving this equation gives

$$T_2 = \frac{1+\varphi(1)}{1+\varphi(1)-\Psi'(1)}T_0 = \frac{1+\varphi(1)}{\varphi(1)-(1-\beta_0)\varphi'(1)}T_0, \tag{57}$$

which has a solution $T_2 > 0$ iff $\varphi(1) > (1-\beta_0)\varphi'(1)$. If $\varphi(1) \leq (1-\beta_0)\varphi'(1)$, $s(t)$ is given by (51) for $t > 0$. Next, $s(t)$ will be derived for $t > T_2$ for the case that $\varphi(1) > (1-\beta_0)\varphi'(1)$. The shock speed $s'(t)$ is given by $s'(t) = \frac{C(t,s(t))}{\Psi(C(t,s(t)))}$. On the shock position, the solution is given by $C(t,s(t)) = (\Psi')^{-1}\left(\frac{t-T_0}{s(t)}\right)$. This gives the following differential equation in $s(t)$:

$$s'(t) = \frac{(\Psi')^{-1}\left(\frac{t-T_0}{s(t)}\right)}{\Psi\left((\Psi')^{-1}\left(\frac{t-T_0}{s(t)}\right)\right)}, \quad s(T_2) = \frac{T_2}{1+\varphi(1)}. \tag{58}$$

Solving this differential equation gives the shock position $s(t)$ for $t > T_2$ for the case that $\varphi(1) > (1-\beta_0)\varphi'(1)$. Summarizing, the following has been found for the shock position $s(t)$:

$$s(t) = \begin{cases} \frac{t}{1+\varphi(1)} & \text{if } \varphi(1) \leq (1-\beta_0)\varphi'(1); \text{ for } t > 0 \\ \frac{t}{1+\varphi(1)} & \text{if } \varphi(1) > (1-\beta_0)\varphi'(1), \text{ for } 0 < t < T_2; \\ \frac{T_2}{1+\varphi(1)} + \int_{T_2}^{t} \frac{(\Psi')^{-1}\left(\frac{\bar{t}-T_0}{s(t)}\right)}{\Psi\left((\Psi')^{-1}\left(\frac{\bar{t}-T_0}{s(t)}\right)\right)} d\bar{t} & \text{if } \varphi(1) > (1-\beta_0)\varphi'(1), \text{ for } t > T_2. \end{cases} \tag{59}$$

Now, the solution for the concentration of suspended bacteria can be constructed:

$$C = \begin{cases} 1 & \text{for } (t,x) \in (0,T_0) \times (0,s(t)) \cup (T_0,T_2) \times (x_R(t),s(t)); \\ 0 & \text{for } (t,x) \in (T_0,\infty) \times (0,x_L(t)) \cup \mathbb{R}^+ \times (s(t),\infty); \\ (\Psi')^{-1}(\frac{t-T_0}{x}) & \text{for } (t,x) \in (T_0,\infty) \times (x_L(t),\min(x_R(t),s(t))). \end{cases} \tag{60}$$

The concentration adsorbed bacteria $\overline{C}$ and fixated bacteria S can be found, using equations (38) and (39).

The preferred result is an homogeneous distribution of bacteria. To achieve that, according to the present model, the following relation should hold:

$$L \leq s(T_2), \tag{61}$$

in which L is the length of the column. Substituting equations (51) and (57) into relation (61) gives the following relation between the length of the column L and the switch time T_0.

$$L \leq \frac{T_0}{\varphi(1) - (1-\beta_0)\varphi'(1)}. \tag{62}$$

3.2 Case study

In this subsection the solution, derived in the last subsection is applied to the Langmuir isotherm. According to [Zheng and Bennett 1995], the Langmuir isotherm is given by

$$\varphi(C) = \frac{\alpha\overline{C_{max}}C}{1+\alpha C}, \tag{63}$$

where the positive constant α is the Langmuir constant and $\overline{C_{max}}$ is the maximum adsorption capacity. Substituting (63) in (49) and (57) gives the following expressions in $\mathbb{R}^+$ for T_1 and T_2:

$$T_1 = \frac{1+\alpha+\alpha\overline{C_{max}}}{\alpha\overline{C_{max}}}T_0; \tag{64}$$

$$T_2 = \frac{\frac{(1+\alpha)^2}{\alpha\overline{C_{max}}} + (1+\alpha)}{\alpha+\beta_0}T_0. \tag{65}$$

The following expression for $s(t)$ is derived:

$$s(t) = \begin{cases} \frac{t}{1+\varphi(1)} & \text{for } t < T_2; \\ \frac{(1+\alpha)T_2}{1+\alpha+\alpha\overline{C_{max}}} + \int_{T_2}^{t} \frac{\sqrt{(1-\beta_0)\alpha\overline{C_{max}}(\bar{t}-T_0)}}{\sqrt{(1-\beta_0)\alpha\overline{C_{max}}(\bar{t}-T_0)}+\alpha\overline{C_{max}}\sqrt{s(\bar{t})-(\bar{t}-T_0)}}d\bar{t} & \text{for } t > T_2. \end{cases} \tag{66}$$

The constant states are located at

$$x_L = \frac{t-T_0}{1+(1-\beta_0)\alpha\overline{C_{max}}}; \tag{67}$$

$$x_R = \frac{t-T_0}{1+\frac{(1-\beta_0)\alpha\overline{C_{max}}}{(1+\alpha)^2}}. \tag{68}$$

The solution for the concentration of suspended bacteria is given by

$$C = \begin{cases} 1 & \text{for } (t,x) \in (0,T_0)\times(0,s(t)) \cup (T_0,T_2)\times(x_R(t),s(t)); \\ 0 & \text{for } (t,x) \in (T_0,\infty)\times(0,x_L(t)) \cup \mathbb{R}^+\times(s(t),\infty); \\ \frac{1}{\alpha}\left(\sqrt{\frac{(1-\beta_0)\alpha\overline{C_{max}}\frac{x}{t-T_0}}{1-\frac{x}{t-T_0}}} - 1\right) & \text{for } (t,x) \in (T_0,\infty)\times(x_L(t),\min(x_R(t),s(t))). \end{cases} \tag{69}$$

Finally, the concentration adsorbed bacteria $\overline{C}$ and fixated bacteria S are given by

$$\overline{C} = (1-\beta_0 c^{fix})\frac{\alpha\overline{C_{max}}C}{1+\alpha C}, \tag{70}$$

$$S = \max_{0\leq\bar{t}\leq t}\left\{\beta_0 c^{fix}\frac{\alpha\overline{C_{max}}C}{1+\alpha C}\right\}. \tag{71}$$

3.3 Numerical Methods

The differential equations for the concentrations of bacteria and fixation fluid are solved by the Standard Galerkin Finite Element Method. First, the weak formulation is derived by multiplication by a test function $\eta \in L^2(\Omega)$ and integration over the domain Ω. As an example, the differential equation for the suspended bacteria (29) is taken. For the adsorption isotherm, the Langmuir isotherm (63) is used. This gives

$$\int_\Omega \frac{\partial C}{\partial t}\eta d\Omega + \int_\Omega \frac{\partial C}{\partial x}\eta d\Omega = \int_\Omega \left(-\theta r_{ads} + \theta r_{des}\right)\eta d\Omega. \tag{72}$$

For the time integration, an implicit scheme is used. That gives the following weak formulations for the urea concentration:

$$\int_\Omega \frac{C^{n+1} - C^n}{\Delta t}\eta d\Omega + \int_\Omega \frac{\partial C^{n+1}}{\partial x}\eta d\Omega = \int_\Omega \left(-\theta r_{ads}^{n+1} + \theta r_{des}^{n+1}\right)\eta d\Omega. \tag{73}$$

This equation holds for all $\eta \in L^2(\Omega)$, which vanish at location of the boundary where C and c^{fix} are prescribed explicitly.

The Newton-Cotes quadrature rules have been used for the development of the element matrices and vectors. For this 1D case, line elements are used. Further more, linear basis functions are taken.

The differential equations for the various concentrations of bacteria are coupled, due to the reaction terms r_{ads} (32), r_{des} (33) and r_{fix} (34). Since these reaction rates are also non linear functions of the concentration suspended bacteria (because of the Langmuir isotherm (63)), Newton's method is used for the differential equations containing these reaction rates. Doing so, the three various concentrations of bacteria come together in one matrix-vector system.

This results in two matrix-vector systems: one for the fixation fluid and one for the three various concentrations of bacteria.

The time span has been divided into equisized discrete time steps. First, the differential equation for the concentration fixation fluid is solved. Next, the solution to the coupled system of differential equations for the various concentrations of bacteria is computed. Some results can be found in the next section.

4 Results

In this section, the analytical solution is visualized for some particular times and locations. For one particular time, we show the comparison between the analytical solution and numerical simulations. The length of the column L has been chosen in such a way that relation (62) holds.

The solution expressed by equations (69), (70) and (71) is visualised in Figure 2 and 3. These figures display the concentration of suspended, adsorbed and fixated bacteria at several times and locations. For this visualisation the following values have been used: $\alpha = 1$, $\beta_0 = 0.5$, $\overline{C_{max}} = 1$, $T_0 = 0.5$ and the length of the column $L = s(T_2)$.

The upper left graph of Figure 2 shows the concentrations at the inlet. The concentration of suspended bacteria C and the concentration fixation fluid c^{fix} at the inflow boundary are prescribed in boundary conditions (27) and (28). From equations (63), (70) and (71) the other concentrations ($\overline{C}$ and S) can be found.

The upper right graph of Figure 2 shows the concentrations at $x = 0.5$m. The front of the fixation fluid starts moving at $t = T_0 = 0.5$h, with velocity $q = 1$m/h. At $t = 1$h the front of the fixation fluid reaches the position $x = 0.5$m. At that time and location, both suspended and temporarily adsorbed bacteria are present. A part of the temporarily adsorbed bacteria is fixated by the fixation fluid. That explains the sudden change in the concentration of temporarily adsorbed bacteria $\overline{C}$. This is also in accordance with equation (70). This equation contains the multiplication factor $(1-\beta_0 c^{fix})$. At $t = 1$h the concentration of fixation fluid changes from 0 into

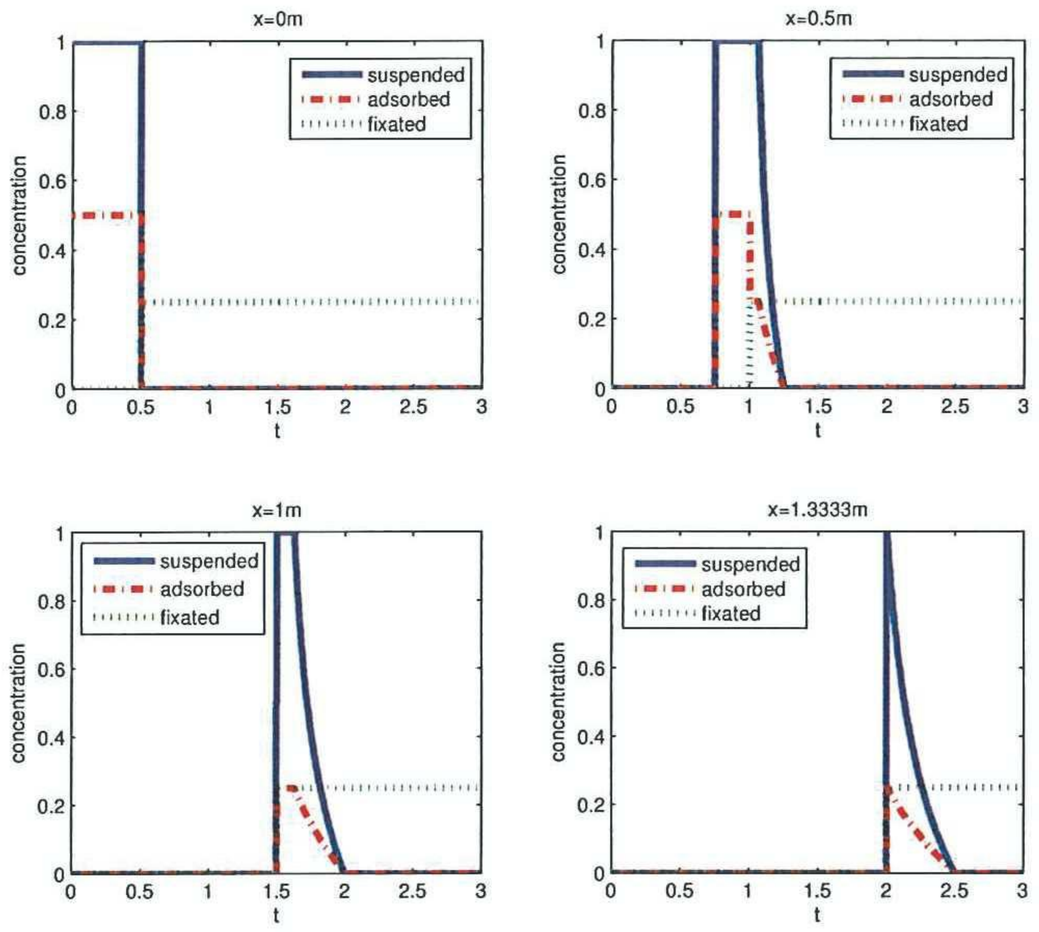

Figure 2: The concentration of suspended, adsorbed and fixated bacteria as a function of time at $x = 0$, 0.5, 1 and 1.3333m.

1 at $x = 0.5$m and consequently the multiplication factor changes from 1 into 0.5. As a result, $\overline{C}$ changes from 0.5 into 0.25. At $t = 1$h, the concentration of fixated bacteria S changes from 0 into 0.25. Since C does not increase in time after $t = 1$h, S does not change anymore.

At $x = 1$m, the pulse bacteria is fully overtaken by the fixation fluid. Hence, the maximum of $\overline{C}$ is 0.25 in the lower graphs of Figure 2. Whereas the graphs in Figure 2 have different maxima for the concentration of temporarily adsorbed bacteria, the maximum of the concentration of fixated bacteria is the same for all the graphs. This has the following reason. When there is no fixation fluid present, the bacteria are not fixated, but as soon as there is fixation fluid present, a part of the bacteria is fixated. According to (71), the maximum depends on the maximum of C, which is in all cases equal to 1.

The lower right graph of Figure 2 shows the concentrations at the outflow boundary.

The upper left graph of Figure 3 shows the initial situation: all concentrations are equal to 0 as prescribed in (26).

The upper right graph of Figure 3 shows the situation in which the pulse bacteria is partly overtaken by the fixation fluid. At $t = 1$h the front of the fixation fluid is located at $x = 0.5$m. The fixation fluid causes a sudden change in the concentration temporarily adsorbed bacteria as was also observed in Figure 2.

At $t = 2$h the front of the bacterial pulse reaches the outflow boundary as is displayed in the lower left graph of Figure 3. The lower right graph of Figure 3 shows the final situation: all non fixated bacteria are flushed out and only the permanently adsorbed bacteria stay in the domain and can provide the hydrolysis of urea.

As a result of the conservation of bacteria, at each time, the number of bacteria in the domain must equal the number of bacteria that flowed in minus the bacteria that flowed out:

$$\int_0^t qC_{in}d\bar{t} - \int_0^t qC_{out}d\bar{t} = \int_\Omega (C + \overline{C} + S)d\Omega, \tag{74}$$

in which C_{in} is the inflow concentration and C_{out} is the concentration at the outflow boundary. This condition holds, as is required.

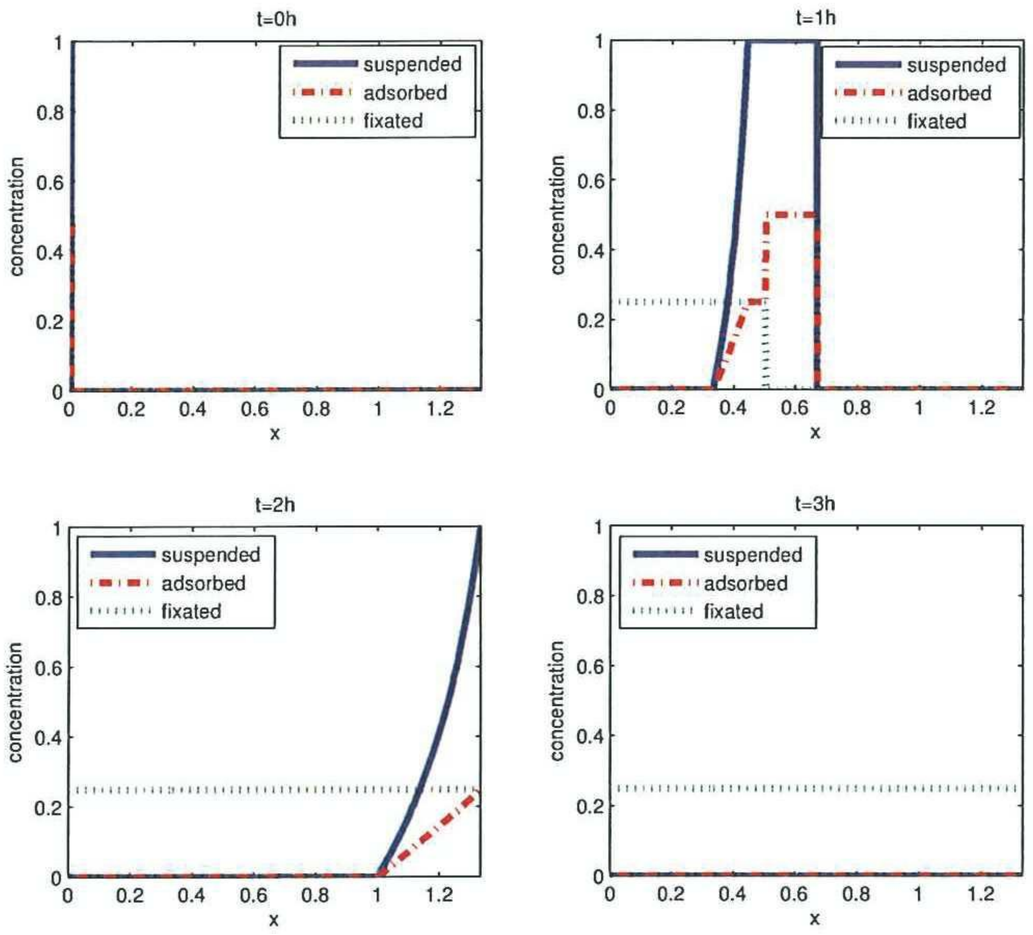

Figure 3: The concentration of suspended, adsorbed and fixated bacteria as a function of location at several times.

Furthermore, the analytical solution is compared with the results of the numerical simulations. This is presented for one particular time: $t = 1h$, but could have been done for any other time. The numerical simulations have been done for several values of the reaction constants k_{ads}, k_{des} and k_{fix}. Figure 4 shows the results of this comparison. As a value for the reaction constants has been chosen: $k_{ads} = k_{des} = k_{fix} = 0.01$, $k_{ads} = k_{des} = k_{fix} = 0.1$, $k_{ads} = k_{des} = k_{fix} = 1$ and $k_{ads} = k_{des} = k_{fix} = 10$. As a time and place step has been chosen: $\Delta x = 0.001$m, $\Delta t = 0.001$h.

From Figure 4 it can be seen that, the larger the reaction constants, the more the numerical solution approaches the analytical solution. There is hardly any difference between the graph for $K = 1$ and the graph for $K = 10$ in Figure 4.

Figure 5 shows the results of refinement. The time and place steps have been decreased two times with a factor 2. It follows that in most cases the numerical solution approaches the analytical solution even more for smaller time and place steps, although it is not a necessarily result of refining. Each step of refining should result in a better approximation of the exact solution and in the limit the numerical solution will equal the exact solution. In this case however, the exact solution is not the analytical (instantaneous) solution but the solution to the model with *finite* reaction rates, with $K = 10$ as a reaction constant. Therefore, the numerical solution with $K = 10$ will not converge to the analytical solution as Δt and Δx tend to zero.

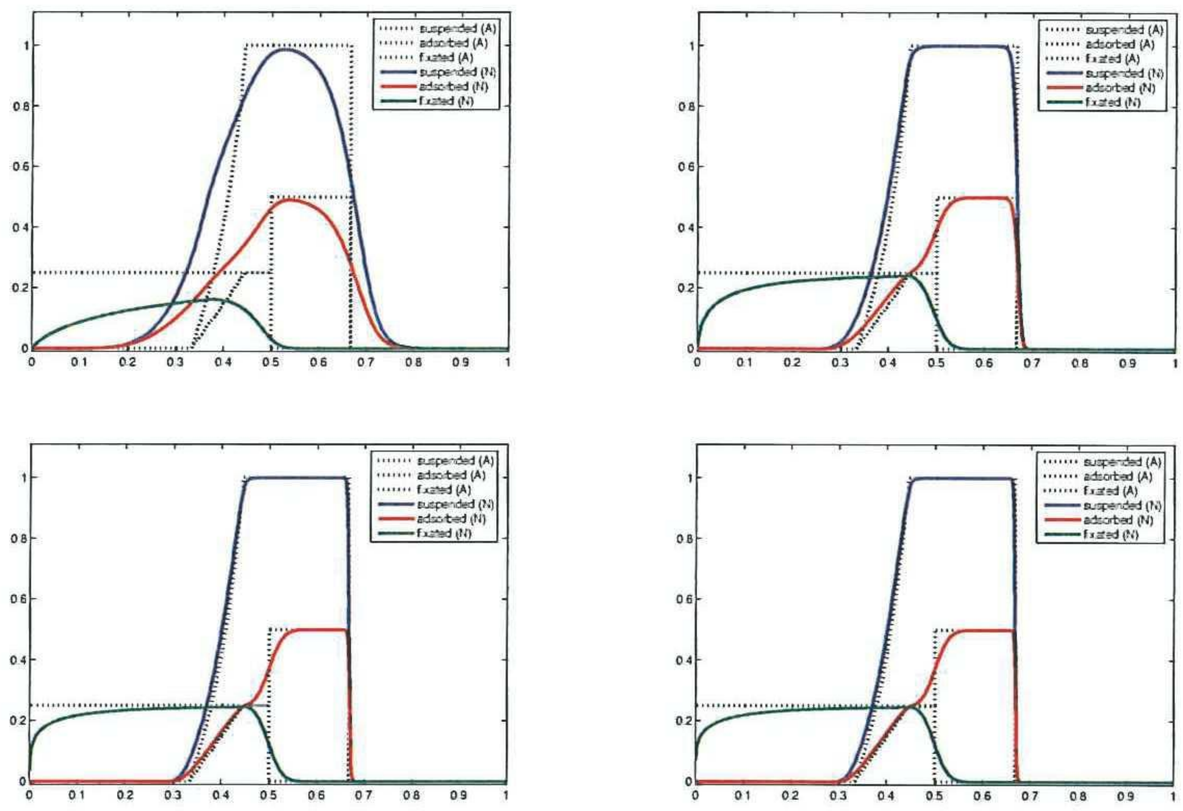

Figure 4: Comparison of the analytical solution (A) at t=1h with the solutions from numerical simulations (N) for several values of the adsorption, desorption and fixation constant ($K = k_{ads} = k_{des} = k_{fix}$). The following values have been assigned to the constants: K=0.01 (top left), K=0.1 (top right), K=1 (bottom left), K=10 (bottom right). The graphs of the analytical solutions are marked with an A and the graphs of the numerical solutions are marked with an N.

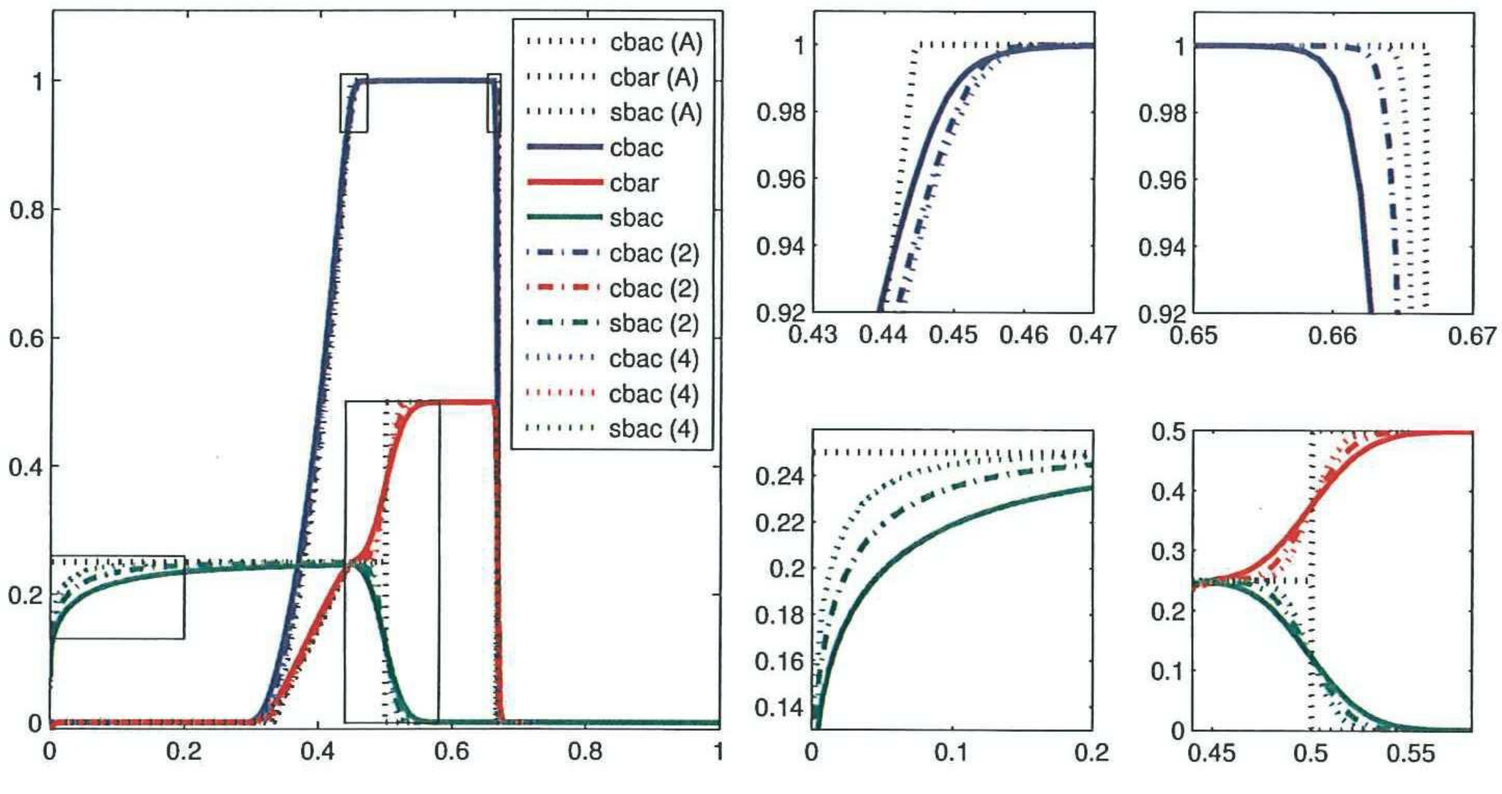

Figure 5: Comparison of the analytical solution at t=1h with the numerical solution for decreasing time and place steps. Some details of the graph in the left subplot are given in the four right subplots. The number between brackets is the refinement factor. Again, the analytical solutions are marked with an A. In the numerical solutions, the reaction constant has been chosen to be $K = 10$.

5 Discussion and Conclusions

A model has been derived for the placement of bacteria. While deriving this model, it turned out that the shock speed needs to be constant until time T_2 to avoid a violation of the conservation of mass principle. To avoid unphysical behaviour, the desorption, adsorption and fixation rate should be chosen carefully.

An analytical solution has been constructed for a simplified version of this model, based on instantaneous equilibria of adsorption, desorption and fixation.

The solutions from the numerical simulations corresponds well with the analytical solution.

The solution can easily be adapted for other values for the porosity θ and the Darcy velocity q.

The simplified model does not contain diffusion or dispersion. Adding these phenomena to the model will result in smoother graphs. The model contains the most important phenomena of the transport of bacteria. Of course, other phenomena can be added, like decay, growth and systematic motion of bacteria. The ratio of the fixated bacteria β versus the adsorbed bacteria depends on the circumstances. Additional research needs to be carried out to find a good expression for this ratio. The values of the various constants in the model should also follow from real life experiments.

From the comparison between the analytical and the numerical solutions the effects from reaction kinetics upon setting small numbers for the reaction constants can be seen. A large reaction constant means that the reaction is fast. If the reaction is very fast compared to the other processes it can be assumed that the equilibrium is instantaneous. From the figures in this paper can be seen that, the larger the reaction constants are, the more the numerical solution approaches the analytical solution, and hence, the instantaneous case.

Refinement of the place and time step results in smaller numerical errors and in most cases better convergence to the analytical solution.

In future, the analytical solution will be compared to real life bacterial placement experiments.

Appendix 1: LIST OF SYMBOLS

C	=	normalized concentration of suspended bacteria, [1];
$C^{adsorbed}$	=	normalized concentration of (temporarily and permanently) adsorbed bacteria, [1];
$\overline{C}$	=	normalized concentration of temporarily adsorbed bacteria, [1];
S	=	normalized concentration of fixated bacteria, [1];
Ψ	=	normalized total concentration of bacteria, [1];
c^{fix}	=	concentration of the fixation fluid, [kmol/m^3];
θ	=	porosity, [1];
$\mathbf{q}$	=	Darcy velocity, [m/h];
$\mathbf{v}$	=	pore water velocity, [m/h];
D_{bac}	=	dispersion/diffusion coefficient for bacteria, [m^2/h];
D_{fix}	=	dispersion/diffusion coefficient for the fixation fluid, [m^2/h];
r_{ads}	=	adsorption reaction rate, [1/h];
r_{des}	=	desorption reaction rate, [1/h];
r_{fix}	=	fixation reaction rate, [1/h];
k_{ads}	=	adsorption rate constant, [1/h];
r_{des}	=	desorption rate constant, [1/h];
k_{fix}	=	fixation rate constant, [1/h];
φ	=	adsorption isotherm;
β	=	factor that describes which part of the adsorbed bacteria are fixated, [1];
β_0	=	factor that describes which part of the adsorbed bacteria are fixated, [m^3/kmol];
α	=	Langmuir constant, [1];
$\overline{C_{max}}$	=	maximum adsorption capacity, [1];
L	=	length of the column, [m];
T_0	=	time at which the injection of bacteria is stopped and the injection of fixation fluid is started, [h];
T_1	=	time at which the front of the fixation fluid reaches the front of the pulse with bacteria, [h];
T_2	=	time at which the shock speed changes, [h].

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