Density-dependent dispersion in heterogeneous porous media Part I: A numerical study

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Abstract

In this paper we describe carefully conducted numerical experiments, in which a dense salt solution vertically displaces fresh water in a stable manner. The two-dimensional porous media are weakly heterogeneous at a small scale. The purpose of these simulations, conducted for a range of density differences, is to obtain accurate concentration profiles that can be used to validate nonlinear models for high-concentration-gradient dispersion. In this part we focus on convergence of the computations, in numerical and statistical sense, to ensure that the uncertainty in the results is small enough.

Concentration variances are computed, which give estimates of the uncertainty in local concentration values. These local variations decrease with increasing density contrast. For tracer transport, obtained longitudinal dispersivities are in accordance with analytical findings. In the case of high density contrasts, stabilizing gravity forces counteract the growth of dispersive fingers, decreasing the effective width of the transition zone. For small log-permeability variances, the decrease of the apparent dispersivity that is found is in agreement with laboratory results for homogeneous columns.

Key words: Heterogeneous porous media, high-concentration-gradient dispersion, brine transport, solute transport, density-dependent flow, macrodispersion, stochastic media, concentration variance

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1 Introduction

1.1 High-concentration-gradient dispersion

This numerical study is part of an investigation of the nonlinear effects that occur when dispersive transport is affected by large density differences. From laboratory experiments it has become clear that the commonly used linear Fickian-type law for dispersion no longer holds when concentration gradients become large. As a result of density gradients, gravity plays a stabilizing or destabilizing role depending on the configuration. In the stable case this results in a reduction of dispersive mixing and smaller effective dispersivities. For a short overview of this topic we refer to Schotting and Landman [1].

The laboratory experiments that were performed on high-concentration-gradient dispersion by Bouhroum [2], Hassanizadeh et al. [3], Moser [4], and Anderson [5] were essentially one-dimensional. In this study we consider the same simple configuration, with unidirectional mean flow. However, on the small scale of heterogeneities the flow is two-dimensional. This is the scale where the interaction between density gradients and gravity is affecting the dispersive spreading. These processes are mimicked in detail in a two-dimensional heterogeneous porous medium. Next, the computed concentrations are averaged over the horizontal (transversal flow) direction. The obtained up-scaled behavior can be described by a one-dimensional model and compared to the various laboratory experiments [2–7]. The study is limited to stable displacements: fresh water is always on top of the heavier salt water solution.

Our purpose is to obtain numerical results that can be used to validate newly developed theories for nonlinear high-concentration-gradient dispersion, such as the one proposed by Hassanizadeh and Leijnse [8]. In this paper we focus on convergence in a numerical and statistical sense, and estimate the uncertainty in the results. Moreover, a comparison with laboratory experiments is made.

1.2 Stochastic transport

We study a range of density differences, from tracer densities up to brine densities as high as 1200 kg/m$^3$. Dispersion of non-reactive tracers has been the subject of many studies. It is common to describe the heterogeneous porous medium in terms of stochastic parameters. Generally, the permeability is assumed to be log-normally distributed, characterized by a mean value and a variance. The correlation function describes how permeability values are spa-
tially correlated, where the correlation scale can be interpreted as a measure of the characteristic size of heterogeneities. However, even in cases where the domain under consideration is large compared to the correlation scale, the uncertainty in predicted concentration values can be very large. Individual porous media replica’s with the same statistical characteristics can generate very different results. Therefore, averaging over an ensemble of realizations is necessary in order to obtain statistically meaningful results.

In the famous work of Gelhar and Axness [9], expressions for macroscale dispersion coefficients are given in terms of the stochastic parameters for both isotropic and anisotropic porous media. Their analysis is valid in the limit of large displacements only, when the scale of the flow system is large compared to the correlation scale. Dagan’s theory [10] is able to describe the evolution of macrodispersivity with time, and for large time approaches the Gelhar and Axness results.

From numerous numerical and theoretical studies on dispersive tracer transport in stochastic media, some conclusions can be drawn that are of interest for this study:

1. Under ergodic conditions, dispersion coefficients tend to constant values after tens of correlation lengths (Dagan [10]). The ergodic hypothesis holds for averaging over large areas compared to the correlation scale, or equivalently over an ensemble of realizations.

2. For single realizations there is no evidence that asymptotic macrodispersivities are reached, even after hundreds of correlation lengths (Trefry et al. [11]). According to Dagan [12], ergodic conditions are eventually reached in the limit $t \to \infty$, but for small initial solute source sizes compared to the correlation scale, travel times need to be extremely large to reach the asymptotic limit.

3. Results differ greatly between individual porous medium realizations having the same stochastic parameters (Smith and Schwartz [13]). Therefore, to make predictions of solute concentrations in field situations (single realizations), not only ensemble averaged results are needed but also a measure of the deviation (uncertainty).

4. The uncertainty in concentration values in single realizations can be very large, and is highest in areas where the concentration gradients are large (Graham and McLaughlin [14], Vomvoris and Gelhar [15], Cvetkovic et al. [16]). The uncertainty decreases with increasing microscale (pore scale) dispersion (Black and Freyberg [17], Graham and McLaughlin [14], Kapoor and Gelhar [18], Dagan and Fiori [19], Andrićević [20]).

5. Where breakthrough is considered at a plane transversal to the flow direction, the uncertainty in the averaged concentration decreases with increasing size of the averaging area (3D) or line (2D) (Black and Freyberg [17], Cvetkovic et al. [16], Dagan and Fiori [19]).
Applying the above to the problem under consideration here, it is not to be expected that single realization results yield constant dispersion coefficients after tens or even hundreds of correlation lengths. Averaging needs to be done over a number of realizations, depending on the width of the column with respect to the correlation length. In order to make predictions for field situations, it is important to know how strongly concentrations in individual realizations (or in the field) may deviate from their ensemble average. Therefore we investigate the topic of concentration variance thoroughly. The relevance of this topic has been emphasized by various authors, e.g. Smith and Schwartz [13], Vomvoris and Gelhar [15], Kapoor and Gelhar [21]. New in the present study is the effect of (stabilizing) density gradients on the concentration variance.

1.3 Outline

The outline of this paper is as follows. In Section 2 the numerical model and its assumptions are presented. Section 3 discusses the numerical convergence and accuracy of the computations, as well as the convergence in a statistical sense. The influence of the density contrast on both types of convergence is discussed. Estimates of the errors caused by the numerical scheme and by the use of a finite number of realizations are given and compared. In Section 4 results are presented, and the differences between tracer and high-density transport are analyzed, for their concentration profiles, concentration variances, and macrodispersivities. For tracer transport, longitudinal macrodispersivities are compared with analytical results. For density-dependent transport a comparison with laboratory experiments is made.

In Part II of this paper a comparison is made between the present numerical results and predictions based on two recently developed nonlinear dispersion models. The objective of the second part is to validate the nonlinear models, and to discuss their applicability and limitations. The effect of various parameters, such as the density contrast, the mean specific discharge, and the log-permeability variance will be investigated further.

2 Numerical model

The main assumption in our analysis is that the standard equations for mass transport in porous media are valid at the local scale, i.e. the scale of heterogeneities. The local scale therefore is already an averaged scale (Darcy scale) and should not be confused with the microscale of individual grains and pores.

The two-dimensional porous media used in this study are heterogeneous only
in the intrinsic permeability, i.e. the porosity is assumed to be constant. The domain under consideration is large compared to the scale of these heterogeneities. Though heterogeneous, the permeability is a scalar, i.e. the medium is assumed to be isotropic. The flow is described by Darcy’s law

$$q = -\frac{k}{\mu} (\nabla p - \rho g),$$  \hspace{1cm} (1)$$

where $q$ is the specific discharge, $k$ the intrinsic permeability, $p$ the fluid pressure, and $g$ the gravity vector.

The viscosity $\mu$ and density $\rho$ of the fluid depend on temperature, on concentration, and to a lesser degree on pressure. In this study isothermal flow is considered, the density being a function of the salt concentration only. For simplicity, viscosity variations are disregarded. We are interested in rotational flow induced by horizontal density gradients. However, in reality horizontal viscosity gradients induce rotational flow as well. The viscosity effect can be neglected when it is small compared to the density effect, which is the case for

$$q_z \ll \frac{k_0 g \rho \gamma_\rho}{\mu \gamma_\mu},$$  \hspace{1cm} (2)$$

where $\gamma_\rho = \partial \ln \rho / \partial c$ and $\gamma_\mu = \partial \ln \mu / \partial c$. With their ratio $\gamma_\rho / \gamma_\mu = 0.37$, the average permeability $k_0 = 5 \cdot 10^{-12}$ m$^2$, $g = 9.81$ m/s$^2$, and $\mu = 10^{-3}$ kg/ms, this implies $q_z \ll 1.83 \cdot 10^{-5}$ m/s. In the simulations presented here a value of $0.6 \cdot 10^{-5}$ m/s is used for the mean specific discharge in the vertical direction.

For a non-adsorbing, non-reactive solute in a non-deformable porous medium, the mass balance is given by

$$n \frac{\partial (\rho c)}{\partial t} + \nabla \cdot (\rho c q - \rho D \nabla c) = 0,$$  \hspace{1cm} (3)$$

where $n$ is the porosity, and $c := \omega / \omega_s$ is a scaled mass fraction, in which $\omega_s$ is the mass fraction of the brine, i.e. the maximum value attained. The scaled mass $c$ fraction therefore lives between 0 (fresh water) and 1 (brine).

Note that we will refer to $c$ as concentration instead of mass fraction. The scaled concentration $C/C_s$ and the scaled mass fraction are identical when tracer transport is considered ($\rho =$ constant, $\omega = C/\rho$). In the literature, tracer transport is commonly discussed in terms of the concentration and the concentration variance. In this paper the same terminology is used, although we realize that for density dependent transport the scaled mass fraction and scaled concentration are not equivalent.
The diffusion/dispersion tensor $D$ is given by (Scheidegger [22])

$$D = (D_m + \alpha_{\perp}q)I + (\alpha_{\parallel} - \alpha_{\perp})qq/q,$$

where $D_m$ is the effective molecular diffusivity, $\alpha_{\perp}$ and $\alpha_{\parallel}$ the transversal and longitudinal dispersivities, $I$ the unit tensor and $q$ the magnitude of the specific discharge vector. Conservation of total fluid mass is described by

$$n \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho q) = 0.$$  (5)

Finally, the equations are coupled by the fluid density, given by the equation of state

$$\rho = \rho_f e^{\gamma_s c},$$

where $\gamma_s = \ln(\rho_s/\rho_f) = \gamma_{\rho} \omega_s$, with $\rho_f$ the density of fresh water, and $\rho_s$ the brine density, corresponding to $c = 1$.

Initially there is a steady flow of fresh water, i.e. $c = 0$ in the column. Then at $t = 0$, salt water (with concentration $c = 1$) enters at the bottom of the column and displaces the resident fluid. The boundary conditions are constant concentrations at the bottom ($c(z = 0) = 1$) and top ($c(z = H) = 0$) of the column, a prescribed pressure at the bottom, and a prescribed discharge $q_0$ at the top. At the vertical boundaries $\nabla c \cdot n = 0$ and $q \cdot n = 0$, i.e. no flow across the boundary. The constant discharge boundary condition is chosen at the top of the column instead of at the bottom. According to Lessoff et al. [23], a constant velocity boundary condition leads to a small decrease of longitudinal dispersion in the vicinity of the boundary. With a constant pressure at the bottom of the column we avoid this effect. Furthermore, in our analysis the brine front is always far enough from the outflow boundary of the column for the results not to be influenced by the particular choice of the boundary conditions.

To simulate the flow and transport the computer code $d^3f$ is used, which was developed to simulate variable density flow in porous media in two- or three-dimensional geometries (Fein, [24]). The program is based on the software package UG (Bastian et al. [25]), a toolbox for discretizing and solving partial differential equations. This package incorporates innovative numerical techniques such as adaptive local grid refinement on unstructured meshes, and robust multigrid methods. An implementation for massively parallel MIMD computers is available. The application for density driven flow in porous media ($d^3f$) has been applied to several model problems, see Johannsen [26,27].
The two-dimensional computational domain is spatially discretized by means of a uniform grid of square cells. The set of equations given above is discretized using a finite volume method with central differences in space. For the time discretization the fractional step $\theta$ scheme (Rannacher [28]) is used with constant time steps. It is second order consistent. For details, see Johannsen et al. [26]. The set of nonlinear algebraic equations is solved using a Newton multigrid method. In our case, the grid refinement is always uniform.

The permeability fields are generated using $FGEN$, a random field generator developed by Robin et al. [29]. We use an isotropic Gaussian log-permeability correlation function $R(h) = \exp(-h/\lambda)^2$. This correlation function has a zero gradient at $h = 0$ and generates smoother fields than the also frequently used exponential correlation function $R(h) = \exp(-h/\lambda)$, which decays more rapidly and has less correlation for small values of the two-point distance $h$. Kapoor and Gelhar [18] recommend not to use the exponential correlation function in numerical modelling for a number of reasons. Moreover, Lessoff et al. [23] state that the Gaussian correlation function yields statistical moments of flow and transport that are representative of other $\ln k$ correlation functions.

<table>
<thead>
<tr>
<th>Porous medium parameters</th>
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<tbody>
<tr>
<td>Height</td>
<td>$H$</td>
<td>1.2</td>
</tr>
<tr>
<td>Width</td>
<td>$W$</td>
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</tr>
<tr>
<td>Correlation length</td>
<td>$\lambda$</td>
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<tr>
<td>Average permeability</td>
<td>$k_0$</td>
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<td>Variance of $\ln k$</td>
<td>$\sigma^2$</td>
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</tr>
<tr>
<td>Effective molecular diffusivity</td>
<td>$D_m$</td>
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</tr>
<tr>
<td>Longitudinal microscale dispersivity</td>
<td>$\alpha_\parallel$</td>
<td>1 \times 10^{-4}</td>
</tr>
<tr>
<td>Transversal microscale dispersivity</td>
<td>$\alpha_\perp$</td>
<td>1 \times 10^{-4}</td>
</tr>
<tr>
<td>Porosity</td>
<td>$n$</td>
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</tr>
</tbody>
</table>

The porous medium characteristics are given in Table 1. Note that the microscale transversal and longitudinal dispersivities are chosen to be equal, i.e. the dispersion tensor is isotropic. Due to the smallness of $\sigma^2$, the microscale dispersion tensor is hardly affected by velocity variations, and is approximately constant. Therefore, the microscale dispersion behaves very similar to molecular diffusion, which is disregarded in the model. This was verified by choosing a molecular diffusion coefficient equal in magnitude to the mean microscale dispersion coefficient (based on the mean specific discharge), and setting the
microscale dispersivities to zero. The results were not significantly different from those based on a velocity-dependent dispersion tensor.

\[
\begin{align*}
\n\n\text{Fig. 1. Permeability field (unit m}^2, \text{dimensions in m), with computed concentration contours (c = 0, 0.1, 0.3, 0.5, 0.7, 0.9), and imposed boundary conditions.}
\end{align*}
\]

An example of a porous medium realization is depicted in Figure 1, with concentration contours computed at the end of a tracer displacement experiment. The permeability varies over about one order of magnitude.

3 Convergence of computations

3.1 Numerical convergence

In this section the numerical convergence of the computations is discussed, and estimates of the discretization errors are given. The two most extreme cases are considered: tracer transport in a homogeneous and in a heterogeneous column. The homogeneous column represents the numerically “worst case”, because the concentration gradients remain large. The only mechanism of spreading in a homogeneous medium is dispersion and/or diffusion at the microscale. In a heterogeneous column, the local variations in permeability cause local variations in velocity. As a result, concentration contours do not remain horizontal, i.e. dispersive fingering occurs (see Figure 1), which increases the effective transition zone width. Therefore, for tracer transport in a heterogeneous medium, the apparent macrodispersivity is much larger than the microscale longitudinal dispersivity.
However, the situation changes drastically when a significant density difference between the two fluids exists. The horizontal density gradients resulting from the variable local velocity field are a driving force for rotational gravity flow. Gravity forces stabilize the front, counteracting the growth of dispersive fingers, and reducing the effective width of the transition zone. In the most extreme case of a very high density contrast and a very small specific discharge (in order to give gravitation effects enough time to stabilize the front), the front behaves as if it is travelling through a homogeneous column instead of through a heterogeneous one. It is this limit for which we first perform the error analysis, by studying the numerical convergence of simulations in a homogeneous column. The advantage of conducting the test for a homogeneous column is that a column with a small width is sufficient, because the flow problem in this case is strictly one-dimensional.

The objective of the numerical convergence study is to determine the grid and time step sizes which are small enough, such that the numerical dispersion is small compared to the microscale dispersion. If this is the case for the homogeneous column, the discretization will surely be accurate enough for the heterogeneous column.

To study the numerical convergence, and to give estimates of the discretization errors, solutions for a number of grid and time step levels are obtained. The main goal of this study is to obtain accurate one-dimensional averaged concentration profiles that can be used for further analysis of nonlinear dispersion theories. These profiles are obtained by averaging the concentrations horizontally over the full width $W$ of the column, i.e.

$$\bar{c}(z, t) := \frac{1}{W} \int_0^W c(x, z, t) dx.$$  \hspace{1cm} (7)

Solutions obtained on subsequent grid levels are compared using the norm

$$||\bar{c}^{j+1,m} - \bar{c}^{j,m}|| := \frac{1}{HT} \int_0^T \int_0^H |\bar{c}^{j+1,m}(z, t) - \bar{c}^{j,m}(z, t)| dz dt,$$  \hspace{1cm} (8)

where $\bar{c}^{j,m}(z, t)$ denotes the horizontally averaged concentration computed on grid level $j$ and time level $m$. Note that we integrate over the full height $H$ of the column, and up to a time $T$ corresponding to a travel distance of 108 correlation lengths. This is the maximum travel distance for which the computed concentrations in any case remain unaffected by the boundary conditions at the top of the column.

The solutions on subsequent time levels are compared in a similar way. In
Each refinement, the grid size or time step size is halved. The result of this investigation is depicted in Figure 2. The numbers in the figure denote the difference between the neighboring solutions in the norm defined in (8).

![Grid Levels and Times](image)

**Fig. 2.** Differences (unit $10^{-3}$) between solutions on subsequent time and grid levels in the norm defined by (8), for tracer transport in a homogeneous column.

By comparing solutions on successive grid and time levels, an estimate of the temporal and spatial discretization errors can be made. When the convergence is monotonic, i.e. for sufficiently fine grids or time steps, Richardson extrapolation can be used, see Ferziger and Perić [30]. The temporal discretization error in $\bar{c}^{10,2}$ is estimated as

$$||\bar{c}^{10,2} - \bar{c}^{10,\infty}|| = \frac{||\bar{c}^{10,2} - \bar{c}^{10,1}||}{2^p - 1} = \frac{1.451 \cdot 10^{-3}}{2^{1.884} - 1} = 5.39 \cdot 10^{-4},$$  

(9)

where the order $p$ is estimated by

$$p = \log \left( \frac{||\bar{c}^{10,1} - \bar{c}^{10,0}||}{||\bar{c}^{10,2} - \bar{c}^{10,1}||} \right) \log 2 = 1.884.$$  

(10)

Similarly, the order of the spatial discretization on time level $m$ is estimated based on the differences between $\bar{c}^{10,m}$, $\bar{c}^{9,m}$, and $\bar{c}^{8,m}$. For $m = 2$ this results in an order of 2.03. The spatial discretization errors are estimated analogous to (9), yielding

$$||\bar{c}^{10,0} - \bar{c}^{\infty,0}|| = 3.91 \cdot 10^{-4},$$

10
\[ ||\bar{c}^{10,1} - \bar{c}^{\infty,1}|| = 3.03 \cdot 10^{-4}, \]
\[ ||\bar{c}^{10,2} - \bar{c}^{\infty,2}|| = 2.38 \cdot 10^{-4}. \]  \hspace{1cm} (11)

The total numerical error on grid level 10 and time level 2 satisfies the inequality

\[ ||\bar{c}^{10,2} - \bar{c}^{\infty,\infty}|| \leq ||\bar{c}^{10,2} - \bar{c}^{10,\infty}|| + ||\bar{c}^{10,\infty} - \bar{c}^{\infty,\infty}||. \]  \hspace{1cm} (12)

The continuous solution \( \bar{c}^{\infty,\infty} \), which would be obtained for infinitely fine grid and time step sizes, is not known. However, the last term in (12) may be estimated by

\[ ||\bar{c}^{10,2} - \bar{c}^{\infty,2}||, \]

because the spatial discretization error is monotonically decreasing with increasing time level (see (11)). The total numerical error is estimated to be

\[ ||\bar{c}^{10,2} - \bar{c}^{\infty,\infty}|| \leq ||\bar{c}^{10,2} - \bar{c}^{10,\infty}|| + ||\bar{c}^{10,\infty} - \bar{c}^{\infty,2}|| = 7.77 \cdot 10^{-4}. \]  \hspace{1cm} (13)

To give an idea of the magnitude of this error, the maximum slope of the computed profiles at long time \( (q_0 t/n = 100 \lambda) \) is compared with the maximum slope of the 1D theoretical long time solution for an infinitely long column:

\[ c = \frac{1}{2} \text{erfc} \left( \frac{z - qt/n}{2\sqrt{\alpha qt/n}} \right). \]  \hspace{1cm} (14)

For the homogeneous column \( (\alpha = \alpha_\parallel = 10^{-4} \text{ m}) \), the relative error in the maximum slope computed on grid level 10 and time level 2 is 1.5%. It is the slope of the profile that is of main interest, as it determines the width of the transition zone between fresh water and brine. The relative error in the slope could be brought back to 0.5% by decreasing the time step size once more. However, as multiple realizations are necessary, and results need to be obtained for a range of density differences and specific discharges, computational time is an important issue. Therefore, we decide to accept the error at grid level 10 and time level 2, and use these levels to perform all high-density computations. This grid level corresponds to 1024 by 2048 grid cells, and the time level to a Courant number of 2.2. With this resolution, one run performed on 16 pentium IV processors takes approximately 20 hours. Note that the homogeneous column is the worst case. In general, concentration gradients in a heterogeneous column will be smaller, leading to better numerical approximations.

Next, the opposite limiting case is discussed: tracer transport in a heterogeneous column. The expected long time macrodispersivity, based on the medium parameters given in Table 1, is an order of magnitude larger than the microscale dispersivity (the only dispersive mechanism in the homogeneous
Fig. 3. Differences (unit $10^{-3}$) between solutions on subsequent time and grid levels in the norm defined by (8), for tracer transport in a heterogeneous column.

Therefore, we expect for tracer transport in a heterogeneous column more dispersed fronts, and as a result, a smaller relative influence of numerical dispersion. To save computational time and to enable us to produce results for a large number of realizations, a coarser grid is preferred for the tracer case. For the heterogeneous tracer case the results of a comparison of different discretization levels is given in Figure 3.

Comparison of the differences in this figure with those in Figure 2 at identical discretization levels, shows that the differences are one order of magnitude smaller for the heterogeneous case. Comparing solutions at different grid levels but equal time levels (going from left to right in Figure 3), shows that the differences become smaller by approximately a factor four in each refinement step. The same factor is found in the time step refinements. This indicates that both the temporal and spatial discretization errors reduce with an order of two. Analogous to (10), using the numbers depicted in Figure 3, the orders for the temporal and spatial discretization in the heterogeneous case are estimated as 1.98 and 1.99 respectively. For both the homogeneous and the heterogeneous case the estimated orders are close to two, as is expected from the discretization schemes. However, the estimations based on Richardson’s method [30] are more accurate in the heterogeneous case than in the homogeneous case. This is because at the same discretization levels, the numerical solution is closer to convergence in the heterogeneous case than in the homogeneous case.

For the heterogeneous case on grid level 9 and time level 2, an upper limit for
the total numerical error $||\bar{c}^{9,2} - \bar{c}^{\infty,\infty}||$ of $1.76 \cdot 10^{-4}$ is estimated from Figure 3. This is even smaller than the error on grid level 10 for the homogeneous case. Grid level 9 ($512 \times 1024$) and time level 2 ($Co = 1$) are satisfactory for the case of tracer transport in the heterogeneous column. Note that on this grid level the grid size is still an order of magnitude smaller than the correlation scale.

3.2 Convergence of ensemble averaging

A criterium is needed in order to decide whether the ensembles of 1-D concentration profiles are converged or not, i.e. that averaging is done over enough realizations. In none of the papers we are familiar with, a clear criterium for this convergence is given. Therefore, we decide to compare results of different ensembles based on the same norm used for comparison of results at different discretization levels. This is an overall measure of the difference in the concentration profiles, as defined by equation (8).

In Figure 4 the difference between ensembles of 10 and of 5 realizations $||\bar{c}_{10} - \bar{c}_{5}||$ (obtained at levels $l = 9$ and $m = 2$) is plotted against the density difference. Also plotted is the difference between solutions obtained on grid levels 9 and 10 (for $m = 2$). This difference is chosen because it is representative for the upper estimate of the total numerical error, for both the homogeneous and the heterogeneous case. For the homogeneous case, the difference is $7.35 \cdot 10^{-4}$ (see Figure 2), and the total error is estimated as $7.77 \cdot 10^{-4}$ (see (13)). Analogously to (13), we obtain $9.2 \cdot 10^{-5}$ for the total error in $\bar{c}^{10,2}$ for the homogeneous case, compared to $8.9 \cdot 10^{-5}$ for the difference between $\bar{c}^{10,2}$ and $\bar{c}^{9,2}$ in Figure 3. We assume that for all intermediate cases of various density contrasts, the total numerical error in $\bar{c}^{10,2}$ can be estimated by the difference between $\bar{c}^{10,2}$ and $\bar{c}^{9,2}$. So, instead of performing the full numerical convergence analysis for every density, the analysis is confined to comparing solutions on grid levels 9 and 10.

Figure 4 shows that the difference between ensembles of 5 and of 10 realizations decreases with increasing density contrast, i.e. increasing the size of the ensemble has little effect for high densities. Due to the stabilizing action of gravity, the effect of the heterogeneities is diminished, as well as the differences between individual realizations. Refining the grid from level 9 ($512 \times 1024$) to level 10 ($1024 \times 2048$) shows the opposite behavior; the difference increases with increasing density contrast as the transition between salt and fresh water becomes sharper.

For density contrasts of 25 kg/m$^3$ and larger there is no need to increase the number of realizations, as the difference between subsequent ensembles is in
the order of or smaller than the numerical error. For the tracer case and density contrasts of 6.25 and 12.5 kg/m$^3$ we decided to perform the simulations on grid level 9, as this has little effect on the solutions and it enables us to do more realizations. For the tracer case the ensemble is extended to 80 realizations, in order to be as accurate as possible in making a comparison with existing stochastic-analytic theories. The difference between 80 and 40 realizations $||\bar{c}_{80} - \bar{c}_{40}||$ is even smaller than the discretization error $||\bar{c}_{9,2} - \bar{c}_{\infty,\infty}||$ for the tracer case, both also plotted in Figure 4.

Note that the numbers presented in Figure 4 for $||\bar{c}_{10} - \bar{c}_{5}||$ depend on the value of the specific discharge. The strength of the gravity effect depends on the ratio of the density contrast and the mean specific discharge. In all simulations the mean discharge was fixed; the highest density contrast of 200 kg/m$^3$ corresponds to a gravity number of 1.64. However, regardless of the value of the specific discharge, the trend shown in Figure 4 is always the same. Figure 4 should be interpreted in a qualitative sense, as the absolute values also depend on the specific choice of the norm (8). Recall that the maximum numerical error found by this norm, corresponds to a 1.5% difference in the maximum slope of the concentration profile at large time.

To further illustrate the magnitude of the differences in Figure 4, averaged concentration profiles over 5 and 10 realizations are plotted in Figure 5 for the two extreme cases. Note that the two profiles for $\Delta \rho = 200$ kg/m$^3$ can hardly be distinguished. Even for the tracer case the difference between ensembles of 5 and 10 realizations is already very small. Note also the difference in steepness between the tracer and high-density profiles, the latter being less dispersed.
4 Results

4.1 Concentration variance

4.1.1 Local concentration and average concentration variances

In our setup, we study the progression of a brine front that covers the full 60 cm width of the column. Thus, concentration averaging is performed over sixty correlation lengths in the horizontal (transversal) direction. According to Bellin et al. [31], who conducted Monte Carlo simulations using one particle per random field, averaging over a number of uncorrelated particle trajectories is equivalent to averaging over the same number of correlation lengths in the transversal flow direction. In their findings, the second-order moments converged after 1500 single particle realizations. Based on this result dispersivities are expected to converge after averaging over 25 realizations.

Before analyzing the second-order moments or dispersivities, first the convergence of the averaged concentration itself will be discussed. To this end, wider but shorter columns were used, with dimensions 4.8 by 0.6 m. In the analysis it is assumed that averaging over ten columns with a width of 4.8 m is equivalent to averaging over one column of 48 m width, i.e. small boundary effects are neglected. In Figure 6 the averaged concentration is plotted against the number of correlation lengths over which is averaged. This is the average concentration computed at a time instant which corresponds to the front having travelled approximately fifty correlation scales. Shown are concentrations at
three vertical positions: close to the middle of the front where $c = 0.5$, and closer to the fringes where approximately $c = 0.1$ and $c = 0.9$.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6}
\caption{Concentration values averaged over $W/\lambda$ transversal correlation lengths}
\end{figure}

As can be seen in Figure 6, the concentrations converge to constant values for large $W/\lambda$. From the results of Bellin et al. [31] we expect convergence after averaging over 1500 correlation lengths. Figure 6 shows that indeed after approximately 1000 correlation lengths the averaged concentrations become almost constant. Note that there is no significant difference in the behavior at the three different cross sections.

Recall that for each realization with index $i$, the concentrations are averaged horizontally over the width $W$ of the column:

$$\bar{c}_i(z, t) := \frac{1}{W} \int_0^W c(x, z, t) dx.$$  \hspace{1cm} (15)

The ensemble average of the concentration is then given by the average over all realizations

$$< c(z, t) > := \frac{1}{N} \sum_{i=1}^{N} \bar{c}_i(z, t).$$  \hspace{1cm} (16)

where $N$ denotes the number of realizations.

The total concentration variance $\sigma_c^2$ at height $z$ and time $t$ (omitted for con-
venience) is defined by

\[
\sigma_c^2 := \langle (c - < c >)^2 \rangle.
\] (17)

Additionally, we define the concentration variance of a single realization \( \sigma_i^2 \) as

\[
\sigma_i^2 := (c - \bar{c}_i)^2,
\] (18)

and the variance of the horizontally averaged concentration \( \sigma_{\bar{c}}^2 \) as

\[
\sigma_{\bar{c}}^2 := \langle (\bar{c}_i - < c >)^2 \rangle.
\] (19)

In the expressions above, the overlying horizontal line denotes averaging by means of horizontal integration, whereas \( < > \) denotes arithmetic averaging over all realizations \( N \).

The total concentration variance \( \sigma_c^2 \) is a measure of the deviations of local concentration values from the ensemble average. The realization variance \( \sigma_i^2 \) expresses the variation of local concentrations with respect to their horizontal average within a single realization. Finally, the variance of the horizontally averaged concentration \( \sigma_{\bar{c}}^2 \) is a measure of the deviations of the horizontally averaged concentrations \( \bar{c}_i \) (of single realizations) from their ensemble average. Combining (17)-(19) yields the following relation between the three variances

\[
\sigma_c^2 = \langle \sigma_i^2 \rangle + \sigma_{\bar{c}}^2.
\] (20)

Of course the third variance depends on the width of the column over which averaging is done. As observed in Figure 6, the average concentration approaches the ensemble average for large averaging lengths, i.e. \( \lim_{W \to \infty} \bar{c}_i = < c > \). Therefore, when the width of the column is very large with respect to the correlation length, the second term on the right hand side of (20) vanishes, i.e. \( \lim_{W \to \infty} \sigma_{\bar{c}}^2 = 0 \), and the concentration variance can be approximated by the average of the variances within individual realizations. We find that for columns of 60 correlation lengths width, the second term in (20) is two orders of magnitude smaller than the first term in (20). For columns with a width of 480 \( \lambda \), the terms differ by more than three orders of magnitude.

For the tracer case simulations are performed for 80 different realizations of the permeability field. Shown in Figure 7 is the ensemble averaged concentration profile at \( q_0 t / n \lambda = 49 \), including the confidence interval based on one standard deviation \( \sigma_{\bar{c}} \). Most of the realizations yield horizontally averaged concentration profiles that lie within this range. The variability of local concentration values (within realizations w.r.t. the horizontal average \( \bar{c}_i \)) is much larger, as shown
Fig. 7. Ensemble averaged concentration profile for the tracer case, with $\sigma_i$ confidence interval, and the average standard deviation of local concentrations $<\sigma_i^2>^{1/2}$.

by the dashed line in Figure 7. The maximum variance is found at the center of the profile, where the concentration gradient attains its maximum value. This is in accordance with findings of Graham and McLaughlin [14], Vomvoris and Gelhar [15], and Cvetkovic et al. [16].

Even though the variance $\sigma_i^2$ is small compared to $<\sigma_i^2>$, there is an interesting feature in $\sigma_i^2$ that we would like to note. In Figure 8, $\sigma_i^2$ is plotted at different time instants. Initially the variance is bimodal, but at some point the two peaks disappear to give an essentially unimodal shape, after which it transforms to bimodal again. In the local concentration variances $\sigma_i^2$ this behavior is not observed, they are always unimodal as is their average in Figure 7. The bimodal-unimodal transition was observed by Andričević [20], who notes that the concentration variance may be constantly bimodal for small enough Péclet numbers. According to Vanderborght [32], the variance is bimodal for low Péclet numbers and sufficiently large travel times. The reason that this bimodal behavior is only observed in the variance of the horizontally averaged concentration and not of the local concentration lies in the effective dispersivity. For local concentrations the Péclet number is too large ($\text{Pe} = \lambda/\alpha_L = 100$), whereas the averaged concentration experiences an effective macrodispersivity that is one order of magnitude larger than the microscale dispersivity, resulting in a smaller Pe.

The realization concentration variance $\sigma_i^2$, which is unimodal, is a measure of the deviations of local concentrations w.r.t. their horizontal average. These local fluctuations are illustrated in Figure 9. Shown is the concentration over a horizontal line at $z = 97.5\lambda$ for one porous medium realization, simulated
Fig. 8. Variance of horizontally averaged concentrations w.r.t the ensemble mean concentration at different travel times for the tracer case.

for $\Delta \rho = 0$ (tracer case) and $\Delta \rho = 200 \text{ kg/m}^3$ (high-density). The dashed and dotted horizontal lines in Figure 9 represent the horizontal average plus/minus one standard deviation $\sigma_i$. The spatial fluctuations in concentration are clearly much larger for the tracer case than for the high-density case. Next, the effect of gravity stabilization on the concentration variability will be discussed.

Fig. 9. Local concentrations at $z = 97.5\lambda$, with one $\sigma_i$ confidence intervals, for $\Delta \rho = 0$ (tracer) and $\Delta \rho = 200 \text{ kg/m}^3$ ($q_0 = 0.6 \cdot 10^{-5}\text{ m/s}$).
4.1.2 Effect of density gradients on concentration variances

When the cases of tracer and brine transport in a perfectly homogeneous column are compared, there is no noticeable difference in the dispersion of the front. In the homogeneous case the concentration contours are perfectly horizontal, the concentration and density gradients are strictly vertical. In this situation no rotational flow is induced, i.e. the velocity is unidirectional. The only difference between the high-density and the tracer in this case is that, using the full continuity equation, in the density-dependent case the velocity is not divergence free. Whereas the ensemble mean front velocity would be uniform over the whole column for a tracer, it is locally increased at the brine front in the density-dependent case. This effect, caused by fluid volume changes, is analyzed for one-dimensional flow by Van Duijn and Schotting [33]. In our numerical results for the homogeneous column, there is no difference in the shape of the concentration profiles with increasing density, but indeed a slight shift due to the increased velocity is found. This is in accordance with the findings of Van Duijn and Schotting [33].

In the heterogeneous case however, permeability variations induce velocity variations, causing the front to lose its initially horizontal shape. Concentration gradients arise in the horizontal direction. With a density contrast, and density gradients no longer being strictly vertical, this results in gravity driven rotational flow. Stabilizing gravity forces counteract the horizontal gradients, forcing the interface to a more horizontal position. This effect is illustrated in Figure 10, where the contour plots of the tracer and of the high-density case are depicted. The tracer concentration contours deviate much more from horizontal lines than the contours in the high-density case. Also the width of the transition zone between brine and fresh water is significantly reduced in the latter case. This is in accordance with Figure 9, showing that local concentration fluctuations over a horizontal line are much smaller for the high-density case than for the tracer case.

![Figure 10](image)

Fig. 10. Computed concentration contours for the tracer and high-density case.

The transition zone width not only depends on the density contrast, but also on the mean velocity of the front. When the velocity is large, gravity has relatively less time to cancel the irregularities in the front, whereas for a very
small velocity it may be able to cancel the horizontal gradients almost completely. The parameter that controls the shape of the computed concentration profiles is the ratio between the density difference and the mean specific discharge. Furthermore, it turned out that our results can be compared to the laboratory experiments of Bouhroum [2] and Moser [4] with the use of the non-dimensional gravity number

\[ N_g = \frac{Ra}{Pe} = \frac{\Delta \rho k_0 g}{\mu q_0}. \] (21)

This number is the ratio between the Raleigh and Péclet numbers, expressing the relative strength of gravity induced flow with respect to forced convection. In Figure 10, the contrast between the tracer and high-density case is large because of the relatively small discharge \((q_0 = 0.6 \cdot 10^{-5} \text{m/s})\), corresponding to an effective front velocity \(q/n\) of 1.3 m/day, and yielding a gravity number of 1.64.

The variance of local concentrations decreases with increasing gravity number (or with the density difference when \(q_0\) and other parameters are kept constant). The variance attains its maximum at the center of the front, recall Figure 7. In Figure 11 this maximum is plotted against travel time, for several density differences. Figure 11 is in a qualitative sense similar to Figure 2 in the paper of Dagan and Fiori [19], which gives the concentration variance at the center of an infinitesimal square plume as a function of time. Dagan and Fiori observe a decrease in concentration variance with increasing microscale transversal dispersion (decreasing \(Pe_{\perp}\)) similar to the decrease we find with increasing density difference (or gravity number). The effect of microscale dispersion on the concentration variance has been discussed in various papers (e.g. Black and Freyberg [17], Graham and McLaughlin [14], Vomvoris and Gelhar [15], Kapoor and Gelhar [18], Dagan and Fiori [19], Andrićević [20], and Vanderborght [32]). Transversal dispersion is a transport mechanism that acts in a way similar to the gravity flow in our problem, acting against horizontal concentration gradients.

The variance of the averaged concentration \(\sigma_c^2\) also decreases with increasing density contrast. Keeping in mind Figure 10, one can imagine that the difference between single realizations in the high-density case is much smaller than for a tracer, and as a consequence less realizations are necessary to obtain convergence of the ensemble average. Recall that this was one of the conclusions in Section 3.
4.2 Longitudinal dispersivities

4.2.1 Tracer dispersion

Next, longitudinal macrodispersivities obtained from tracer simulations are compared with an existing analytical solution. Dispersivities are obtained using two different methods: from the central second order moment (of the concentration gradient), and by fitting with the long-time analytical solution of the linear advection-dispersion equation (14). See for details of these two methods the Appendix.

In Figure 12 the macrodispersivities (microscale dispersion is eliminated) are compared to the analytical solution of Hsu [34], who bases his work on that of Dagan [10,12]. Hsu gives expressions for macrodispersivities as a function of travel distance in two-dimensional isotropic media with various ln$k$ correlation functions, amongst which is the Gaussian function used in the present numerical study. Taking into account that the correlation scale $\lambda$ in our definition is equal to $\sqrt{\pi/2}$ times the integral scale, Hsu’s local or time-dependent dispersivity as a function of travelled correlation lengths $z/\lambda$ for a Gaussian correlation function is

$$\frac{\alpha}{\lambda \sigma^2} = \frac{1}{4} \left[ -1 + \frac{2(z/\lambda)^2}{(z/\lambda)^3} \exp \left[ -(z/\lambda)^2 \right] + \frac{1}{2} \frac{3(z/\lambda)^2}{(z/\lambda)^3} + 2 \sqrt{\pi} \text{erf}(z/\lambda) \right]$$

(22)

where the position $z$ is equivalent with $q_0t/n$. However, even for the ensemble...
of 80 realizations we are not able to reproduce the local dispersion coefficient. Note that with local dispersivity the local macroscale dispersivity is addressed, and not the microscale dispersivity. The computed local dispersivity oscillates around the analytical solution of Hsu. This local dispersivity is derived from a comparison of profiles at subsequent time steps, see the Appendix. However, when the concentration profiles are fitted with with the erfc-solution (14), the obtained dispersivity is the result of the front having travelled through the column up to a certain point (or equivalent time). In other words, the apparent dispersivity is an effective average over the travelled length. Therefore, this apparent dispersivity can be compared to the dispersivity obtained from the second moment only if the latter is integrated in time. Then the difference between the two methods is small, see Figure 12.

The integrated solution of Hsu can be approximated, for travel distances over three correlation lengths, by

\[
\frac{\alpha}{\lambda \sigma^2} = \frac{-1}{8(z/\lambda)^3} - \frac{3 \ln(z/\lambda)}{4z/\lambda} + \frac{\sqrt{\pi}}{2} \text{erf}(z/\lambda),
\]  

and is also plotted in Figure 12. For travel distances larger than ten correlation lengths a satisfactory agreement is found between our results and (23), our dispersivities being only a few percent lower. The small discrepancy could be explained by the fact that in Hsu’s analysis microscale dispersion is neglected, while this is present in the simulations. With Pe = 100 we are just at the lower bound of the range for which it is generally accepted that microscale dispersion can be neglected. According to Burr et al. [35], disregarding microscale
dispersion leads to an overprediction of macrodispersivities.

Another thing that Burr et al. [35] note is that in most theoretical studies the second moments are taken with respect to the ensemble mean position, whereas the center position differs in each realization. When dispersivities are obtained with respect to the individual center positions, their ensemble result is smaller. However, our results plotted in Figure 12 were obtained with respect to the ensemble mean center positions, so this can not cause the discrepancy. Furthermore, in our case, where horizontally averaged concentrations are considered, the variance in the centroid positions turned out to have an insignificant effect.

4.2.2 Density-dependent dispersion

Dispersivities were also determined from the numerical results for higher densities. Although the approach as described in the Appendix is only valid for the tracer case where density gradients do not play any role, the same erfc-fitting procedure is used for the density-dependent case. This is done in order to make a comparison with the experimental results by Bouhroum [2], Moser [4], and Jiao and Hötzl [7], who all determine the dispersion coefficients by fitting breakthrough curves with the erfc-solution (14). The apparent dispersivity can be interpreted as a measure of the transition zone width.

![Graph showing macrodispersivities as a function of travel time for various density contrasts.](image)

Fig. 13. Apparent macrodispersivities fitted with the erfc-solution (14) as a function of travel time, for various density contrasts (in kg/m$^3$).

In Figure 13 the fitted macrodispersivity is plotted as a function of travel time for a number of density differences. Except for the tracer case, where 80
realizations were averaged, these results are averages over 10 realizations. After 20 correlation lengths, the dispersivity is practically constant for all density contrasts, except for the tracer dispersivity which still increases slightly. Note that the theoretical asymptotic value for the tracer case is $\sqrt{\pi/2} = 0.886$. The long-time asymptotic dispersivity decreases with increasing density difference (or gravity number).

The decrease in the long-time dispersivity with gravity number is compared to the results of Bouhroum [2], Moser [4], Jiao and Hötzl [7], and Kretz et al. [6]. Moser [4,36] performed four series of laboratory column displacement experiments: low (tracer) concentrations, high absolute concentrations, constant density but variable flow rate, and constant flow rate but variable density. For each experiment we calculated the gravity number, as given in (21), based on the fluid densities and flow rates given by Moser [4]. Even for the low concentration experiments the density effect can be large when the flow rate is small. The set of experiments with high absolute concentrations was performed with two fluids of high concentration, differing slightly in density. These experiments proofed that not the absolute density, but the difference in density between the fluids is of importance.

The experiments of Bouhroum [2] were similar to Moser’s experiments, and cover a wide range of gravity numbers. The gravity numbers computed from the data of Jiao and Hötzl [7] are all but one in the tracer range. These three studies were all performed in essentially homogeneous porous media. Kretz et al. [6] however, used two different heterogeneous media, consisting of different arrangements of blocks of glass beads of various sizes.

In Figure 14, the experimental dispersivities are plotted against the corresponding gravity number, together with our numerical results. The dispersivities are scaled by the tracer dispersivity. In our case the tracer dispersivity is known from the numerical experiments with $\Delta \rho = 0$. In the laboratory experiments there is always a slight density difference, so the tracer dispersivity is approximated by the value obtained from the experiment with the lowest gravity number (Moser [4]: $N_g = 0.0018$). This scaling is necessary because it is the relative decrease in dispersivity that is of interest here. The absolute dispersivities can not be compared, because they depend on the specific porous medium used ($\lambda, \sigma^2$, correlation function). However, Kretz et al. [6] state that the relative decrease in dispersivity does not depend on the medium characteristics. The results depicted in Figure 14 contradict this statement. A satisfactory agreement is found between the experimental data for the different homogeneous media and the simulation results for $\sigma^2 = 0.1$. However, the data for the two heterogenous media (Kretz et al. [6]) show a less strong decrease in dispersivity with $N_g$. The simulation results for the permeability fields with stronger heterogeneity $\sigma^2 = 0.5$ are closer to these results. Apparently, the decrease of $\alpha/\alpha_{tr}$ depends on the log-permeability variance $\sigma^2$. 
Fig. 14. Scaled macrodispersivity against gravity number. Comparison of experimental and simulation results (obtained at $q_0 t / n \lambda = 97.2$). Filled symbols: homogeneous media; open symbols: heterogeneous media.

This result is plausible, because the more heterogeneous the medium, the larger the absolute values of the macrodispersivities. As a result of a stronger spreading in more heterogeneous media, the macroscopic density gradients are smaller than in a less heterogeneous media. So, even for the same gravity number, the density gradients differ for different media. Therefore, the gravity effects in a medium with larger $\sigma^2$ are relatively smaller. Apparently, even for the scaled dispersivity $\alpha/\alpha_{tr}$ this results in a slower reduction with gravity number for larger $\sigma^2$. The experimental results of Bouhroum [2], Moser [4], Jiao and Hötzl [7], and the simulation results for $\sigma^2 = 0.1$ are alike because the tracer dispersivities are all in the order of a millimeter.

The fact that the macrodispersivity is a function of the gravity number, and thus of fluid and flow parameters, is in contradiction with dispersivity being a medium parameter. The linear dispersion model, using a fixed dispersivity, can only model the numerical and laboratory experiments for the tracer case, i.e. when $\Delta \rho = 0$. When high density gradients are present, this approach is no longer adequate (this work, Hassanizadeh and Leijnse [8], Schotting et al. [36], and Anderson [5]). A different approach is proposed by Hassanizadeh and Leijnse [8], who introduce a nonlinear dispersion equation. This equation contains an additional parameter (besides the classical tracer dispersivity), with which experiments for any density contrast can be modelled. In part II of this paper we will discuss the comparison between our numerical experiments and Hassanizadeh and Leijnse’s model, as well as a recently developed nonlinear macroscopic model based on homogenization of the local scale equations.
5 Conclusions

This numerical study confirms that stabilizing gravity forces are responsible for the reduction in the transition zone width between brine and fresh water during column displacement experiments. Concentration profiles, obtained by averaging the local concentrations over horizontal lines, become steeper as the density contrast between the fluids increases. The parameter that controls the relative strength of the density effect as opposed to forced convection is the gravity number \( N_g = Ra/Pe \). The simulations for the log-permeability variance of \( \sigma^2 = 0.1 \) resemble the behavior observed in homogeneous media. The decrease of the scaled macrodispersivity with gravity number is found to be in satisfactory agreement with the experimental results of Bouhroum [2] and Moser [4]. However, the relation between the scaled macrodispersivity and the gravity number depends on the magnitude of the heterogeneity, i.e. on \( \sigma^2 \).

In heterogeneous media, the local fluctuations in concentration decrease in size with increasing gravity number. This behavior is similar to what is found for an increase in the microscale dispersivity (Dagan [19]). Like microscale transversal dispersion, the gravity forces counteract horizontal concentration/density gradients. The variance in the averaged concentration (of a single realization with respect to the ensemble mean concentration) also becomes smaller for higher density contrasts. As a result, in the case of high density differences less realizations are needed to reach ensemble convergence.

For the tracer case, reasonably good agreement is found between dispersivities obtained from the computed ensemble averaged profiles and the analytical result of Hsu [34], when a time-average of the dispersivity is compared. The time-dependent dispersivity obtained using the second central moments fluctuates around Hsu’s analytical solution.

The simulations for the log-permeability variance of \( \sigma^2 = 0.1 \) resemble the behavior observed in essentially homogeneous porous media. The decrease of the scaled macrodispersivity with gravity number is found to be in satisfactory agreement with the experimental results of Bouhroum [2] and Moser [4]. However, the relation between the scaled macrodispersivity and the gravity number depends on the magnitude of the heterogeneity, i.e. on \( \sigma^2 \).

A thorough investigation was performed to ensure that the numerical and statistical uncertainties in the results are small and of the same order. The computed profiles can be used to test the validity of nonlinear dispersion models applicable for high-concentration-gradient dispersion.
Appendix. Determination of apparent longitudinal dispersivity

In this appendix two methods for determining the dispersivity from the horizontally averaged concentration profiles are explained. Both approaches are valid only for tracer concentrations, because they rely on the linear one-dimensional convection-diffusion equation. This equation is obtained from the more general solute transport equation (3) written in one-dimensional form with the assumption of a constant density. Using a coordinate system moving with the mean front velocity $q_0/n$, i.e. $x^* := x - q_0t/n$, we obtain (dropping the asterisk)

$$\frac{\partial c}{\partial t} = D_l(t) \frac{\partial^2 c}{\partial x^2},$$

(24)

where the local (time-dependent) dispersion coefficient $D_l$ is assumed independent of $x$.

The method of moments is commonly used to determine dispersion coefficients in studies of travelling plumes. In our study of an interface between brine and fresh water we apply the same method, but slightly altered. Instead of taking the second moment of the concentration itself, we use the gradient of concentration. The concentration gradient in our case is similar in shape and behaves like the concentration distribution of a travelling plume, originating from an initial pulse input and vanishing at the domain boundaries. For the gradient of concentration $c' = \partial c/\partial x$ the same transport equation holds

$$\frac{\partial c'}{\partial t} = D_l(t) \frac{\partial^2 c'}{\partial x^2}.$$

(25)

Multiplication by $x^2$ and integration over space yields

$$\int_{-\infty}^{\infty} \frac{\partial c'}{\partial t} x^2 dx = D_l(t) \int_{-\infty}^{\infty} x^2 d \left( \frac{\partial c'}{\partial x} \right).$$

(26)

Performing integration by parts twice to the right hand side integral and using the fact that $c'' = c' = 0$ at $x = \pm \infty$ gives

$$\frac{\partial}{\partial t} \left( \int_{-\infty}^{\infty} c' x^2 dx \right) = -D_l(t) \int_{-\infty}^{\infty} \frac{\partial c'}{\partial x} 2xdx = 2D_l(t) \int_{-\infty}^{\infty} c' dx.$$

(27)
Finally, using the boundary conditions $c = 1$ at $x = -\infty$ and $c = 0$ at $x = \infty$, the local dispersion coefficient is given by

$$D_l(t) = -\frac{1}{2} \frac{\partial}{\partial t} I_{xx},$$

(28)

where $I_{xx}$ is the central second moment of the concentration gradient $c'$

$$I_{xx} = \int_{-\infty}^{\infty} c' x^2 \, dx.$$

(29)

The central second moment $I_{xx}$ is computed from the concentration profile at every time step. Its time derivative is approximated by taking the difference on two subsequent times and dividing by the time step size. The local dispersion coefficient $D_l(t)$ computed by (28) is the factor that determines the change of the concentration profile from one time to the next. However, one profile at a certain time $t$ is the result of travelling through the column up to a certain point $q_0 t/n$ (in the original coordinate). During its travel the profile at time $t$ has experienced an average dispersion coefficient given by

$$D_{ave}(t) = \frac{1}{t} \int_{0}^{t} D_l(t) \, dt.$$

(30)

This average dispersion coefficient can be compared to the dispersion coefficient determined by another method. This second method fits the concentration profile at time $t$ with the analytical long time solution of (24), where $D$ is assumed to be constant in space and time:

$$c = \frac{1}{2} \text{erfc} \left( \frac{x}{2\sqrt{Dt}} \right).$$

(31)

The apparent dispersivity $\alpha$ can be obtained by dividing the fitted dispersion coefficient by the front velocity $q_0/n$, when molecular diffusion is negligible. This dispersivity contains the combined effect of microscale dispersion and the variable velocity field. To obtain the macrodispersivity only, the microscale dispersivity is subtracted.

Finally, we note that in experimental studies dispersivities are often obtained by fitting the breakthrough curve (measured at a certain position $L$) with (31), where $x$ is substituted by $L - q_0 t/n$. A three point fitting method as described in Bear [37] is often sufficient. Fitting the concentration profile yields similar results as fitting the breakthrough curve, but is more exact. Fitting
the breakthrough curve introduces a small error because $t$ in the denominator of (31) varies along the breakthrough curve while it is assumed to be constant.

References


