Density-dependent dispersion in heterogeneous porous media Part II: Comparison with nonlinear theories

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Abstract

In this paper, a comparison is given between the results of a series of high-resolution numerical experiments and two nonlinear theories for high-concentration-gradient dispersion. The purpose of this paper is to validate these nonlinear theories and to discuss their limitations. The first theory, introduced by Hassanizadeh and Leijnse, is a modification of Fick’s law which involves a second order term in the dispersive flux equation and an additional dispersion parameter. The numerical experiments confirm the dependency of this parameter on the flow rate that was observed earlier in laboratory experiments. In addition, a dependency on traveled distance is observed, adding to the non-uniqueness of the nonlinear dispersion parameter.

The second theory is based on homogenization of the local scale equations describing density-dependent transport. Gravitationally stable brine dispersion in a weakly heterogeneous porous medium is considered. The homogenization approach leads to a nonlinear system of macroscale equations, in which the dispersion coefficient depends on the dimensionless density gradient. In addition, an expression for the concentration variance is derived. For small log-permeability variances, the homogenization model predictions are in satisfactory agreement with the numerical experiments without the introduction of new (fitting) parameters.

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

Numerous experimentalists [1,3], Hassanizadeh et al. [4], Kempers and Haas [5], Moser [6,7], Anderson [8], Watson et al. [9,10], Kretz et al. [11] have studied the effect of high concentration and density gradients on hydrodynamic dispersion in porous media. The question of interest is whether Darcy’s law for flow through a porous medium and linear Fick’s law for hydrodynamic dispersion remain valid under high-concentration (gradient) conditions. The motivation for most of this research lies in the fact that underground salt formations have long been considered as potential storage sites for radioactive and other hazardous wastes [12]. In the vicinity of deep salt rock formations, highly concentrated brines and large density gradients are found (Visser [13], Frape et al. [14], Hazzanizadeh and Leijnse [15]). With the greatest risk of these storage sites being the escape of radionuclides and transport by groundwater (Roxburg [16], Sander and Herbert [17]), brine transport modelling is an essential element in risk-assessment studies (Warnecke et al. [18]). Furthermore, brine transport modelling is important for salt water intrusion problems.

1.1 Experimental and theoretical findings

From several independent laboratory studies of stable brine displacements in vertical columns it is concluded that linear Fick’s law is inadequate for modelling the experiments beyond tracer concentrations (see e.g. Hassanizadeh and Leijnse [15], Schotting et al. [7], and Watson et al. [9]). However, other researchers still find the linear law applicable, but only when the value of the dispersivity is fitted to the experiments (see e.g. Bouhroum [3], or Jiao and Hötzl [19]). The observed dispersivity depends on both the density contrast between the fluids, and the mean flow rate in the column, and therefore varies from experiment to experiment. This is in contradiction with dispersivity being a porous medium property.

Instead of adjusting the dispersivity value to be in accordance with the experiments, Hassanizadeh and Leijnse [15] introduce a new theory with which the experiments for the whole range of concentration differences is satisfactorily modelled. Their nonlinear equation for the dispersive mass flux $\mathbf{J}$ contains an additional dispersion parameter $\beta$:

$$\begin{equation}
(1 + \beta |\mathbf{J}|)\mathbf{J} = -\rho \mathbf{D} \cdot \nabla \omega.
\end{equation}$$

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In this expression, $|J|$ is the magnitude of the dispersive mass flux vector, $\rho$ is the fluid density, $\omega$ is the salt mass fraction and $D$ is the traditional (tracer) dispersion tensor. Note that linear Fick’s law is obtained in the limit of small dispersive mass fluxes, and, moreover, when $\beta$ is set to zero.

Hassanizadeh and Leijnse [15] show that their nonlinear model adequately describes the experiments of Hassanizadeh et al. [4]. Schotting et al. [7] use the nonlinear model to predict the breakthrough curves obtained by Moser [6]. With the data of Moser [6] it is shown that the dispersion parameter $\beta$ is inversely proportional to the flow rate to some power (Schotting et al. [7]). The flow rate dependency is confirmed by Watson et al. [9]. Her experiments also show that Darcy’s law remains valid under high-concentration-gradient conditions [10].

Whereas Hassanizadeh and Leijnse [15] focus on homogeneous porous media, Welty and Gelhar [20] study the effect of both density and viscosity gradients on dispersion in heterogeneous media. Welty and Gelhar [20] use a stochastic approach, in which the heterogeneous medium is modelled as a random field characterized by statistical parameters. They obtain the traditional macroscopic transport equation, in which the macrodispersive flux is described analogous to Fick’s law. The dispersivity, however, is not a constant, but depends on the concentration gradient. Welty and Gelhar [20] present an approximate explicit expression for the dispersivity in the steady-state limit, for the case of a three-dimensional porous medium with an exponential correlation function.

Even though Welty and Gelhar’s theory [20] applies to heterogeneous media, their observations are very similar to those in homogeneous media. For gravitationally stable displacements, the dispersivity decreases with increasing density difference between the two fluids. Furthermore, this effect is reduced when the mean flow rate is increased. The physical process responsible is the stabilizing effect of gravity, induced by horizontal density gradients that arise as a result of local velocity variations.

Hassanizadeh and Leijnse [15] claim that their observations cannot be due to heterogeneities, and they explain the reduction in dispersivity as a result of an increasing resistance to solute dispersion with an increasing concentration gradient. However, we believe that the reduction in dispersivity is a result of the same process seen in heterogeneous media, but only present at a smaller scale. The velocity dependence of $\beta$ also points in this direction. It implies the existence of two time-scales in the column experiments: the time-scale of the action of gravity, and the convection time-scale, associated with the mean specific discharge in the column, see Schotting et al [7].

Porous media that are considered to be homogeneous are still heterogeneous at the microscale, where velocity variations cause hydrodynamic dispersion.
It is at this scale of heterogeneity that horizontal density gradients arise that interact with gravity forces. In their 1963 review of diffusion and dispersion in porous media, Perkins and Johnston [21] already recognize the suppression of dispersion by gravity on both the scale of individual pores and the scale of permeability heterogeneities. This explains why the observations of Welty and Gelhar [20] are very similar to those of Hassanizadeh and Leijnse [15]. Furthermore, the results of the numerical experiments presented in Part I of this study, show that the decrease in dispersivity for weakly heterogeneous porous media is very similar to that observed in the laboratory experiments in homogeneous media.

Note that in the numerical experiments, the only mechanism that can be responsible for the suppression of dispersion is gravity. The numerical model is based on the assumption of the validity of Darcy’s law and Fick’s law at the local scale, i.e. the scale of permeability heterogeneities. Microscale processes are only accounted for in an averaged sense by the local dispersion tensor. Furthermore, it is verified that setting the gravitational acceleration to zero in a high-concentration experiment, results in concentration distributions identical in shape to those in a tracer experiment. The only difference with the tracer case is a slight shift of the concentration distribution, due to non-Boussinesq volume effects, which are discussed for 1-D brine transport by see Van Duijn and Schotting [22,23]).

1.2 Outline and objectives

The results of the high-resolution numerical experiments described in Part I—in terms of ensemble averaged concentration profiles—are compared to results of three different nonlinear macroscopic dispersion models. The objective of this chapter is to compare the predictive capacity of these nonlinear models, and to discuss their applicability and limitations.

The first model is the nonlinear model of Hassanizadeh and Leijnse [15]. This model has been tested against various laboratory experiments in essentially homogeneous media, whereas the numerical simulations are performed in heterogeneous media. However, the two-dimensional media used in the numerical experiments are only weakly heterogeneous, characterized by small values of the log-permeability variance $\sigma^2$ and the correlation scale $\lambda$. The results in Part I show that the numerical experiments are comparable to the laboratory experiments in homogeneous porous columns, in the sense that the observed dispersivities are of the same order, and show a similar decrease with increasing strength of the density effect. Therefore, we expect that the nonlinear model of Hassanizadeh and Leijnse [15] can be applied here as well. As opposed to the laboratory experiments, the numerical experiments produce concentration
data at many time instants, and in addition, information about the concentration variances. Results are obtained for a range of density contrasts and various fixed mean flow rates, so that the relation between Hassanizadeh’s nonlinear dispersion parameter $\beta$ and these variables can be established.

The second and third dispersion models are based on the application of homogenization theory by Egorov and Demidov, respectively. Egorov et al. [2] have published results for the case of a layered medium. The work discussed here is yet unpublished. Egorov starts from the same balance equations at the local scale as are solved in the numerical simulations. With homogenization (a mathematical upscaling technique), a macroscopic model is derived. Predictions based on this model are compared to the ensemble averaged concentration profiles. The homogenization approach is in many ways similar to the stochastic approach of [20], and leads—with the same assumptions—to the same result. However, [20] study a three-dimensional porous medium with a different type of correlation function, and their model therefore is not directly comparable to the numerical experiments. Moreover, the homogenization approach of Egorov yields an expression for the concentration variance, which can be compared to the variances obtained in the numerical experiments. In this chapter the main steps in the homogenization procedure are explained, while details of the derivation and solution of the so-called cell problem are given in Appendix A.

Finally, Demidov applies the homogenization approach to density-dependent flow at the pore scale level, considering Stokes flow around micro-heterogeneities. Demidov’s analysis leads to results that are comparable to the experiments in essentially homogeneous porous media. Since the numerical simulations for $\sigma^2 = 0.1$ show enough similarity to these type of laboratory experiments, a comparison with Demidov’s model predictions is also made.

2 The nonlinear model of Hassanizadeh

2.1 Numerical implementation

In this section, predictions based on Hassanizadeh’s nonlinear model are used for comparison with the numerical experiments described in Part I. On the averaged (macroscopic) scale, the flow system is one-dimensional. The governing equations in a non-deformable porous medium are given by the fluid mass balance

$$n \frac{\partial \rho}{\partial t} + \frac{\partial}{\partial z}(\rho q) = 0,$$

(2)
and the salt mass balance

\[ n \frac{\partial \rho \omega}{\partial t} + \frac{\partial}{\partial z} (\rho \omega q + J) = 0, \]

(3)

where, following Hassanizadeh and Leijnse [15], the dispersive mass flux \( J \) is given by the one-dimensional version of (1):

\[ (1 + \beta |J|) J = -\rho D \frac{\partial \omega}{\partial z}. \]

(4)

In the above equations, \( n \) denotes the porosity (assumed constant), \( t \) denotes time, \( \rho \) denotes the fluid density, and \( q \) denotes the specific discharge in the \( z \)-direction (positive pointing upward). The dispersion coefficient is given by

\[ D = n d_{mol} + \alpha_{tr} |q|, \]

(5)

in which \( d_{mol} \) is the molecular diffusion coefficient, \( \alpha_{tr} \) is the longitudinal tracer dispersivity, and \( |q| \) is the magnitude of the specific discharge. Finally, the equation of state relates the fluid density to the mass fraction of dissolved solute:

\[ \rho = \rho_0 e^{\gamma \omega}, \]

(6)

where \( \rho_0 \) is the density of fresh water, and \( \gamma_\rho = 0.692 \) is a curve fitting constant.

With the exponential form of the equation of state, the mass fraction in (3) and (4) can be eliminated conveniently using the fluid mass balance (2), yielding

\[ n \frac{\partial \rho}{\partial t} + q \frac{\partial \rho}{\partial z} + \gamma_\rho \frac{\partial J}{\partial z} = 0, \]

(7)

and

\[ J (1 + \beta |J|) = -D \frac{1}{\gamma_\rho} \frac{\partial \rho}{\partial z}. \]

(8)

Next, the density is scaled using

\[ \rho = \frac{\rho - \rho_0}{\rho_s - \rho_0}, \]

(9)

where \( \rho_s \) denotes the salt water (brine) density. Finally, the salt mass balance reads

\[ n \varepsilon \frac{\partial \rho}{\partial t} + \varepsilon q \frac{\partial \rho}{\partial z} + \gamma_\rho \frac{\partial J}{\rho_0 \partial z} = 0, \]

(10)

where the parameter \( \varepsilon \) denotes the relative density difference

\[ \varepsilon = \frac{\rho_s - \rho_0}{\rho_0}. \]

(11)

The dispersive mass flux \( J \) can be solved from (8), yielding for the case of stable displacements, i.e. \( J > 0 \):

\[ J = \frac{-1 + \sqrt{1 - \frac{P \rho_0}{\rho_s \partial z}}}{2\beta}, \]

(12)
with \( P = 4\beta D\varepsilon\rho_t/\gamma\rho \). Combining the salt mass balance (7) and the fluid mass balance (2), results in the following differential equation for \( q \):

\[
\frac{\partial q}{\partial z} = \frac{\gamma\rho}{(\varepsilon\rho + 1)\rho_t} \frac{\partial J}{\partial z}.
\]

The nonlinear system given by (10), (12), and (13) cannot be solved analytically (unless some simplifying assumptions are made, see Schotting et al [7]). Therefore, the system is solved numerically using an explicit finite difference scheme. The boundary conditions are

\[
q = q_0 \quad \text{at} \quad z = H,
\]

\[
\rho = \begin{cases} 
0 & \text{at} \quad z = H, \\
1 & \text{at} \quad z = 0.
\end{cases}
\]

The constant velocity boundary condition is chosen at the end \( z = H \) rather than at the start of the domain \( z = 0 \), to be in accordance with the boundary conditions of the numerical experiments, see Part I. However, the height \( H \) is chosen to be 1.5 times the height of the column in the numerical experiments. The step function in the initial density distribution is approximated by a steep error function, which is centered far enough from the inlet boundary in order to avoid boundary effects. Moreover, it is verified that the steepness of the initial profile is large enough, i.e. a further increase of the steepness has no effect on the computed profiles. Also, the solutions are numerically converged, i.e. further refinement of the grid and time step has no influence on the solutions.

The optimal value of the parameter \( \beta \) is determined using an iterative procedure, based on Newton’s method, which converges rapidly. Here, the optimal value is the value that yields a maximum gradient in the computed density profile equal to that in the experimental profile under consideration. The maximum-gradient fitting is preferred over a least-squares fitting, because it is independent of the precise position of the profile, which depends on the density gradient (as a result of volume effects) and on \( \beta \) itself. Moreover, the maximum gradient is of the highest interest, as it determines the width of the transition zone.

Note that the profiles in the numerical experiments are computed in terms of a scaled mass fraction \( (\omega/\omega_\omega) \) instead of the scaled density, see Part I. For small enough mass fractions \( (\rho_\omega \leq 1025 \text{ kg/m}^3) \), the two are identical because the relation between the density and the mass fraction is almost linear, i.e.

\[
\rho = \rho_t e^{\gamma\rho\omega} \approx \rho_t (1 + \gamma\rho\omega).
\]

However, for larger densities there is a slight difference between the two scaled variables. Therefore, the experimental profiles are converted to scaled density profiles before comparison with the nonlinear model.
2.2 Results

Based on fitting the nonlinear model to the experiments of Moser [6], Schotting et al. [7] conclude that the nonlinear dispersion parameter $\beta$ is only dependent on the flow rate, not on the density gradient. This means that, for a certain medium and flow rate, experiments with different density contrasts can be described using the same value of $\beta$. Watson et al. [9] conclude that $\beta$ decreases with increasing density and viscosity contrasts, although it is unclear on which result this conclusion is based. Furthermore, they show that $\beta$ varies slightly between the different types of sand used.

The three parameters that vary in the numerical experiments are: the density contrast between fresh water and brine $\Delta \rho$, the mean specific discharge $q_0$, and the log-permeability variance $\sigma^2$. Using the numerical results, the dependence of $\beta$ on these three parameters is investigated. Simulations have been carried out for two types of permeability fields: for $\sigma^2 = 0.1$ and $\sigma^2 = 0.5$ respectively. The asymptotic value of the tracer dispersivity $\alpha_{tr}$ is linearly dependent on $\sigma^2$. The magnitude of permeability variations is therefore expressed in the nonlinear macroscopic model by $\alpha_{tr}$. Like in the numerical experiments, molecular diffusion is disregarded so that $D = \alpha_{tr}|q_0|$. The solutions of the nonlinear model depend on four parameters: $\varepsilon$ (determined by $\Delta \rho$), $q_0$, $\alpha_{tr}$, and $\beta$.

Furthermore, by fitting density profiles obtained at different times, the dependence of $\beta$ on travel-time (or corresponding distance) is investigated. This dependency has not been studied before. Both Schotting et al. [7] and Watson et al. [9] use only the measured profiles at the end of the column to determine the value of $\beta$.

The fitted values of $\beta$ can be extremely sensitive to a change in the desired maximum slope in the density profile. This sensitivity is discussed first, and it is used to estimate the uncertainties in the fitted $\beta$ values.

2.2.1 Uncertainty in $\beta$

Running the model for a certain set of parameters ($\varepsilon$, $q_0$, and $\alpha_{tr}$) with an increased value of $\beta$ always results in steeper density profiles. This can be explained as follows. The effect of the nonlinear term in (4) is to decrease the dispersive flux, with respect to the flux given by linear Fick’s law. According to (12), the dispersive mass flux $J$ for a certain gradient $\frac{\partial \rho}{\partial z}$ decreases with increasing $\beta$: $J \sim \beta^{-1/2}$ when $-P \frac{\partial \rho}{\partial z} \gg 1$. A decrease in the dispersive flux has a slowing effect on the smearing of the density profiles, i.e. the profiles remain steeper.

The value of $\beta$ is adjusted until the maximum gradient of the computed density
profile matches that of the experimental profile. Using the value of the two last iterations, the sensitivity of the computed maximum gradient to the value of $\beta$ is determined. This sensitivity largely decreases with decreasing density contrast. In other words, for small density differences, a large change in $\beta$ has little effect on the computed density gradient. This creates an uncertainty in the $\beta$-fitting procedure.

Figure 1 presents the sensitivity of $\beta$ with respect to a variation in the maximum gradient $\gamma_{\text{max}}$. Here, $\gamma$ is the dimensionless density gradient, defined by

$$\gamma = -\frac{g k_0 \frac{\pi^2}{4} \Delta \rho \partial \rho}{\mu \alpha_{\perp} q_0 \partial z},$$

(16)

where $g$ is the acceleration of gravity, $k_0$ the average permeability, $\mu$ the fluid viscosity, and $\alpha_{\perp}$ the local transversal dispersivity. Note that $\rho$ in (16) is the scaled density, whereas the other parameters are dimensional. The dimensionless density gradient is introduced here, because it is the main parameter in the homogenization model, to which comparisons will be made later. As shown in

![Fig. 1. Sensitivity of the nonlinear dispersion parameter](image)

Figure 1, the sensitivity of $\beta$ decreases linearly with $\Delta \rho$ on logarithmic scales. This implies that especially for the numerical experiments with small density contrasts, a small uncertainty in the gradient of the experimental profile results in a relatively large uncertainty in the fitted $\beta$. As a consequence, it is virtually impossible to determine $\beta$ for a perfect tracer experiment ($\varepsilon \downarrow 0$).

The nonlinear model parameter $\beta$ is fitted such that the model predictions match the numerical experiments, for a fixed mean specific discharge ($q_0 = 6.0 \cdot 10^{-6}$ m/s), and a range of density contrasts from 6.25 to 200 kg/m$^3$. The uncertainty in the fitted $\beta$ has three sources. The first two sources are
related to the accuracy of the "experimental" profile, of which the maximum gradient is used to determine $\beta$. The uncertainty in the experimental profile (and its derivative) is determined by the use of a finite number of realizations, and by discretization errors in the numerical model. Both of these sources of inaccuracy are discussed in Part I. The third source of inaccuracy is caused by numerical errors in the macroscopic nonlinear dispersion model. However, because the nonlinear model is only one-dimensional, grid and time step refinement is not a limiting factor. It is verified that this third source of inaccuracy always has the smallest contribution to the total uncertainty in $\beta$.

![Figure 2](image_url)

Fig. 2. Dependency of $\beta$ on the density contrast. Fitted values for $\sigma^2 = 0.1$ and 0.5 at $q_0 t/n\lambda = 100$ and 90, respectively ($q_0 = 0.6 \cdot 10^{-5}$ m/s).

In Figure 2, the fitted values of $\beta$ are plotted for each density, with the corresponding uncertainties. For the medium with $\sigma^2 = 0.1$, the fitting is most accurate for the largest density contrast $\Delta \rho = 200$ kg/m$^3$. However, the uncertainties for all densities are of similar magnitude. For the high-density case, the uncertainty is dominated by the numerical error in the experimental density profile. The uncertainty in the maximum gradient $\gamma_{\text{max}}$ is computed based on the results obtained at different grid and time step levels. See for a discussion of this method Part I. The sensitivity as plotted in Figure 1 is in turn used to compute the corresponding uncertainty in $\beta$.

For small $\Delta \rho$, the effect of using an average over a limited number of 10 realizations is the greatest cause of uncertainty. When it is assumed that the uncertainty in the gradient of the experimental profiles decreases with the square root of the number of realizations, an estimate of the uncertainty can be made based on the difference between the ensembles of 5 and 10 realizations. However, this difference is determined by chance, i.e. other ensembles
of $N = 5$ and $N = 10$ may be more or less alike. Especially for small $\Delta \rho$, this approximation may give an underestimation of the uncertainty.

Considering the results for $\sigma^2 = 0.1$ in Figure 2, the value of $\beta$ is increasing with $\Delta \rho$, except for the value obtained for $\Delta \rho = 200$. The present results seem to be in contradiction with the conclusion of Schotting et al. [7] that $\beta$ is independent of $\Delta \rho$. However, all other ranges show some extent of overlap with the range determined for the highest density, as depicted with the dotted lines in Figure 2. Therefore, none of the other values is conflicting with the value determined for $\Delta \rho = 200 \text{ kg/m}^3$. Moreover, the uncertainty for low values of $\Delta \rho$ may still be underestimated. Even though the uncertainties plotted in Figure 2 are quite large (up to almost 40% for $\Delta \rho = 6.25 \text{ kg/m}^3$), they correspond to variations in the maximum gradient of approximately 5% at most.

On the other hand, the results obtained for $\sigma^2 = 0.5$ exhibit a more monotonic trend. Figure 2 shows a decrease in $\beta$ with decreasing $\Delta \rho$, tending to a constant value. Only the result obtained for the highest density shows overlap with the result for $\sigma^2 = 0.1$. All other $\beta$ values are clearly lower for the higher variance, though still of the same order of magnitude. Thus, as shown experimentally by Watson et al. [9], $\beta$ is medium-dependent.

\[ \beta(\rho) \]

Fig. 3. Dependency of $\beta$ on travel-time. Solid lines: variable $q_0$ (in $10^{-5} \text{ m/s}$); other data: $q_0 = 0.6 \cdot 10^{-5} \text{ m/s}$. 


During the numerical experiments, concentration profiles T are computed at various time instants. One after the other, these profiles are used to fit $\beta$, in order to obtain the dependence of $\beta$ on travel-time. The travel-time is expressed in non-dimensional form by the number of correlation lengths travelled, i.e. $q_0 t/n\lambda$. In Figure 3, the results are depicted for five different flow rates, for a fixed density difference of $200 \text{ kg/m}^3$, and the medium with the smallest permeability variations ($\sigma^2 = 0.1$). The data points are connected by solid lines, all showing a slow increase of $\beta$ with time. A fit through the values for $q_0 = 0.6 \cdot 10^{-5} \text{ m/s}$ is depicted by the dashed line. Based on this fit, the value obtained at a distance of $100\lambda$ is only 42% of the asymptotic long-time value. It is expected that, after $1500\lambda$, $\beta$ reaches 90%, and only after $3500\lambda$, $\beta$ reaches 95% of its asymptotic value.

The values obtained for $\Delta \rho = 100$ and $\Delta \rho = 50 \text{ kg/m}^3$ and flow rate $q_0 = 0.6 \cdot 10^{-5} \text{ m/s}$ are depicted by circles in Figure 3. These points are all close to the data for $\Delta \rho = 200$. The three points obtained at $q_0 t/n\lambda = 100$ are identical to those plotted in Figure 2. Note that the differences plotted on a linear $\beta$-scale in Figure 2 are small relative to the change in $\beta$ with flow rate.

Additionally, Figure 3 shows the data for the same flow rate but stronger heterogeneity ($\sigma^2 = 0.5$), for density contrasts of 200 and 100 kg/m$^3$ respectively. These two cases, depicted with filled and open squares in Figure 3, yield significantly different $\beta$ values. As shown in Figure 2, $\beta$ decreases with decreasing $\Delta \rho$.

Furthermore, note that in Figure 3 the data points for $\sigma^2 = 0.5$ show larger variations than the data for $\sigma^2 = 0.1$. This is due to the limited number of realizations, and the slower convergence to ensemble statistics for the fields with higher variability. Especially at early times, when the brine front has travelled relatively few correlation lengths, the conditions for ergodicity are not fulfilled yet. Also with an increase in flow rate the lines plotted in Figure 3 become less smooth (compare $q_0 = 0.3$ to $q_0 = 4.8$). A smaller effect of gravity stabilization for higher flow rates results in more variability from realization to realization, and therefore a need for a larger number of realizations. See Part I for an extensive discussion of this topic.

Finally, note that the last data point for the highest variance was taken at $90\lambda$ rather than at $100\lambda$. This is because of the larger spreading, causing an earlier influence of the top boundary. However, the difference in $\beta$ at these two time instants is only a few percent, and therefore insignificant in Figure 2.
2.2.3 Flow rate dependence

Figure 3 shows that $\beta$ decreases with flow rate. In Figure 4, this flow rate dependence is plotted on logarithmic scales. The $\beta$ values plotted are obtained at the end of the column (at $q_0 t/n\lambda = 100$ and 90, for $\sigma^2 = 0.1$ and 0.5 respectively). Watson et al. [9], and Schotting et al. [7], observe that, in log-log space, $\beta$ decreases linearly with $q_0$. This indicates that $\beta$ is inversely proportional to the flow rate to some power. The two lines in Figure 4 depict the best linear fit through the data for the two media.

For the weakly heterogeneous permeability fields, a power of $1.85 \pm 0.13$ is found. This is in good agreement with the findings of Watson et al. [9], and Schotting et al. [7], who observe powers of 1.85 (coarse sand) and 1.76 respectively. The result for the fields with stronger heterogeneity, however, show a stronger dependence on flow rate than the (nearly) homogeneous media.

2.2.4 Comparison of profiles

In Figure 5, three density profiles are plotted: the "experimental" profile, i.e. the ensemble average of all numerical experiments, the profile computed with the linear dispersion model, and the profile computed with the nonlinear dispersion model based on Hassanizadeh and Leijnse’s theory [15]. Figure 5a displays the tracer case, i.e. for $\varepsilon = 0$ (or in the nonlinear model $\varepsilon = 10^{-8}$). For all times, both the linear and nonlinear model show an excellent agreement with the numerical experiment. In Figure 5b the high-density case of $\varepsilon = 0.2$ is depicted. The linear model leads to profiles as dispersed as in the tracer case in Figure 5a. The experimental profiles for $\varepsilon = 0.2$ are much steeper, and
better approximated with the nonlinear model. At short time the agreement is less good, because the value $\beta = 5 \cdot 10^5$ used to generate all profiles in Figure 5 is the value fitted for the profile at the end of the column.

For the linear model predictions depicted in Figure 5, the tracer dispersivity $\alpha_{tr}$ is used. Alternatively, the linear model can be used with an adapted dispersivity, fitted to the specific experiment. As described in Part I, one way of determining the apparent dispersivity $\alpha_{app}$ is to fit the experimental profile with the error-function solution of the linear (tracer) dispersion equation. When this "apparent" dispersivity is used—instead of the tracer dispersivity—obviously

![Graphs showing density profiles for different conditions](image-url)

**Fig. 5.** Density profiles of the numerical experiment for $\sigma^2 = 0.1$, $q_0 = 0.6 \cdot 10^{-5}$ m/s, with linear and nonlinear model predictions. (a) tracer case; (b) high-density case.
a better fit is obtained with the linear model. This is shown in Figure 6, which depicts the long-time profiles for $\varepsilon = 0.2$ for both cases. The gradient of the profile computed using $\alpha = \alpha_{\text{app}}$ agrees well with that of the experimental profile. However, at both tails the linear prediction deviates from the experimental curve, which shows more dispersion.

The prediction of the nonlinear model matches the experiment very well for $\rho \leq 0.5$, but is more dispersed for large $\rho$. In fact, the linear and nonlinear solutions are practically rotation-symmetrical around $\rho = 0.5$, but the experimental profile is not. At the brine side the simulated profile is steeper than at the fresh water side. A possible explanation for this asymmetry is that the concentration distribution at the brine side has experienced less heterogeneities due to the smaller travel-time (less correlation scales). In addition, stabilizing density effects, which inhibit dispersion, are stronger for higher concentrations.

In Figure 7, the predictions of the nonlinear model are compared for the two media with $\sigma^2 = 0.1$ and $\sigma^2 = 0.5$. For the high-density experiment ($\varepsilon = 0.2$), both cases are modelled using the same value of $\beta$, see Figure 7a. The model prediction for $\sigma^2 = 0.5$ is not as good as for $\sigma^2 = 0.1$, showing deviations from the experimental curve at the tails. However, the maximum gradient is still satisfactorily approximated. Note that the best fit is obtained for $\beta = 4.3 \cdot 10^5$.

Figure 7b shows the intermediate case of $\varepsilon = 0.025$. The nonlinear model prediction using $\beta = 5 \cdot 10^5$ is much steeper than the experimental profile for the case $\sigma^2 = 0.5$, whereas a good match is found for $\sigma^2 = 0.1$. For $\sigma^2 = 0.5$, a
much better agreement between model prediction and experiment is obtained using the smaller value $\beta = 1 \cdot 10^5$. Recall Figure 2, which shows that for $\sigma^2 = 0.5$ the fitted $\beta$ is significantly smaller for $\Delta \rho = 25$ than for $\Delta \rho = 200$ kg/m$^3$. For $\sigma^2 = 0.1$, experiments for all densities can be satisfactorily modelled using the same $\beta$, i.e. $\beta = 5 \cdot 10^5$. Apparently, for media with stronger heterogeneity this is not possible, and fitting is necessary for every individual experiment.

![Graph](image)

Fig. 7. Experimental profiles compared to nonlinear model predictions for $q_0 = 0.6 \cdot 10^{-5}$ m/s, $\sigma^2 = 0.1$ and $\sigma^2 = 0.5$. (a) high-density case; (b) medium-density case.
3 Homogenization

In this section, the homogenization model is discussed and its predictions are compared to the numerical experiments. Homogenization theory is applied to the case of brine dispersion in weakly heterogeneous porous media. Starting with the local scale equations in two dimensions, a one-dimensional macroscopic model is derived. In this section, the main steps in the homogenization procedure are discussed and the final results are given: a macroscopic balance equation and a rheological relation between the dispersive mass flux and the density gradient. Next, this macroscopic model is used to predict the averaged concentration profiles of the numerical experiments. Comparisons are made for various $\Delta \rho$ and $q_0$ values, and for media with log-permeability variances $\sigma^2 = 0.1$ and 0.5 respectively.

3.1 Derivation of the macroscale model

3.1.1 Problem definition

We consider brine displacing fresh water in a two-dimensional non-deformable porous medium, where the mean flow is in vertical direction. As in the numerical experiments, it is assumed that the flow can be described by Darcy’s law, and the local dispersion by the classical linear relation (Fick’s law). In addition, the Boussinesq approximation is adopted, yielding for the fluid mass balance

$$\nabla \cdot q = 0,$$

where $q$ is the specific discharge or Darcy velocity vector, with components $q_1$ and $q_2$ in vertical and horizontal direction respectively. With the use of the exponential equation of state (6), the salt mass balance written in terms of the density is given by

$$n \frac{\partial \rho}{\partial t} + q \cdot \nabla \rho = d_\parallel \frac{\partial^2 \rho}{\partial x_1^2} + d_\perp \frac{\partial^2 \rho}{\partial x_2^2},$$

where $d_\parallel$ and $d_\perp$ are the longitudinal and transversal components of the local dispersion tensor respectively. It is convenient to use the rotation of Darcy’s law

$$\frac{\partial}{\partial x_2} \left( \frac{\mu q_1}{k} + \rho g \right) = \frac{\partial}{\partial x_1} \left( \frac{\mu q_2}{k} \right),$$

where $x_1$ and $x_2$ are the two spatial coordinates in vertical and horizontal direction respectively, $\mu$ is the dynamic fluid viscosity, and $k$ is the intrinsic permeability. It is assumed that $\mu$, $n$, $g$, $d_\parallel$, and $d_\perp$ are given constants, while the permeability $k$ is a function of spatial coordinates.
Permeability distributions in the field resemble log-normal distributions. Therefore, in stochastic subsurface hydrology, it is common to deal with log-permeability fields, see e.g. Dagan [24] or Gelhar [25]. The permeability is represented as 

\[ k = k_0 \exp(\sigma K), \]

where \( k_0 \) is the average permeability, and it is assumed that \( \sigma \) is small and \( K(x) \) is an isotropic homogeneous random field with zero mean and unity variance. The dimensionless log-permeability \( K \) is assumed to be spatially correlated, by definition of a proper correlation function

\[ R(r) = \langle K(x')K(x'') \rangle, \quad r = |x' - x''|, \]

where the angle brackets denote average quantities. Here, spatial averaging is applied, which for stationary random fields—according to the principle of ergodicity—is equivalent to ensemble averaging over all possible realizations.

Generally, the term \( \nabla \cdot d_{ij} \nabla \rho \) appears on the right-hand side of (18), where the local dispersion tensor \( d_{ij} \) following Scheidegger [26] reads

\[ d_{ij} = (nd_{mol} + \alpha_\perp |q|) \delta_{ij} + \left( \alpha_\parallel - \alpha_\perp \right) \frac{q_i q_j}{|q|}, \]

(21)

Here, \( \alpha_\parallel \) and \( \alpha_\perp \) are the longitudinal and transversal dispersivities correspondingly, \( \delta_{ij} \) denotes the Kronecker delta, and \( d_{mol} \) is the molecular diffusion coefficient. The dispersion tensor \( d_{ij} \) in principle varies in space, due to the local fluctuations in the velocity field. These fluctuations are proportional to the fluctuations in the permeability field, i.e. \( q_1 = q_0 + O(\sigma) \), \( q_2 = O(\sigma) \). Therefore, when \( \sigma \) is small it can be assumed that the local dispersion is proportional to the mean velocity \( q_0 \) rather than to the local velocity. Then, the local dispersion can be described by the right-hand side of (18), provided that \( d_\perp \) and \( d_\parallel \) are defined as

\[ d_\perp = nd_{mol} + \alpha_\perp |q_0|, \quad d_\parallel = nd_{mol} + \alpha_\parallel |q_0|. \]

(22)

The assumption of a constant viscosity seems in contradiction with the fact that the fluid viscosity varies more strongly with concentration than the fluid density, see e.g. Holzbacher [27]. However, we are interested in the vorticity (rotational flow) that is induced by viscosity or density gradients. The viscosity effect is small compared to the density effect when the mean discharge in the vertical direction satisfies

\[ |q_0| \ll \frac{k_0 \rho_0 g \gamma_\rho}{\mu \gamma_\mu}, \]

(23)

where the parameters \( \gamma_\rho \) and \( \gamma_\mu \) describe the relative change in density and viscosity as a result of concentration variations, i.e. \( \partial \ln \rho / \partial \omega \) and \( \partial \ln \mu / \partial \omega \) respectively. The ratio of these two parameters for Sodium Chloride (NaCl) solutions is approximately 2.7. For the low velocities naturally occurring in groundwater, criterion (23) is satisfied and viscosity effects can be disregarded. The dominance of the density effect is observed by Welty and Gelhar [20], and also in the experiments of Krupp and Elrick [28] and of Starr and Parlange [29].
Equations (17)–(19) form a coupled system of equations that must be solved simultaneously in order to describe the flow and density fields. Here, we are concerned with the evolution of an initially horizontal front between fresh water and brine, moving in vertical direction. So it is assumed that the brine density, the mean vertical flow rate, and the mean transverse pressure derivative attain constant values as $x_1 \to \pm \infty$:

$$x_1 \to \pm \infty : \quad \rho = \rho_{\pm}, \quad \langle q_1 \rangle = q_0, \quad \langle e^{-\sigma K q_2} \rangle = 0. \quad (24)$$

Two totally different schemes of front evolution are possible, depending on the sign of $\Delta \rho = \rho_- - \rho_+$. In one case the front is stable, while in the other case it is not and gravitational fingering may occur. This work is restricted to the gravitationally stable case, in which fresh water is on top of the heavier brine, i.e. $\Delta \rho = \rho_s - \rho_f > 0$.

3.1.2 Dimensionless variables

The main goal is to derive a macroscale equation describing the long-time behavior of the dispersion of the brine front. To start, all variables are rendered dimensionless, using characteristic spatial and temporal scales. There are two spatial scales in the problem: the local scale and the macroscopic scale. The first is determined by the heterogeneity size, a measure of the distance over which the permeability is correlated. The common way to define this local scale is to use the integral scale

$$l = \int_0^{\infty} R(r) \, dr. \quad (25)$$

Note that so far, the correlation scale $\lambda$ has been used to characterize the size of the heterogeneities. The numerical model described in Part I of our paper uses the Gaussian correlation function $R(r) = \exp(-h^2/\lambda^2)$. In this definition, however, $\lambda$ is not equivalent with the integral scale $l$; their relation is given by $l = \lambda \sqrt{\pi}/2$. In the homogenization procedure, $l$ will be referred to as the "microscale". However, when comparisons to the numerical experiments are made, the correlation scale $\lambda$ is still used in order to be consistent with the terminology of the preceding parts.

The second scale (macroscale) is given by the width $L$ of the brine front. A basic assumption in this analysis is that the microscale and macroscale are "well separated" i.e. $\varepsilon_x = l/L \ll 1$ (see also Lunati et al. [30]). This assumption is directly related to the objective of describing the long-time behavior. The width of the front increases with time due to dispersive spreading and therefore at large enough time the assumption $\varepsilon_x \ll 1$ is valid independent of the width of the initial density profile.
Two spatial scales produce two types of dimensionless spatial coordinates, according to the general approach of the homogenization theory, see e.g. Bakhvalov and Panasenko [31], Sanches [32], or Bensoussan et al. [33]. The first type (so called "fast" coordinates) is related to the microscale while the second type ("slow" coordinates) is related to the macroscale. The macroscale process is one-dimensional and only dependent on the slow coordinate. It is convenient to use a moving frame macroscale coordinate. So we introduce 

$$\tilde{x}_1 = \frac{x_1}{l}, \quad \tilde{x}_2 = \frac{x_2}{l}, \quad z = \frac{x_1 - q_0 t/n}{L},$$

(26)

as the two fast coordinates and the slow coordinate respectively. The velocity and density fields will be considered as two-scale functions to investigate the transport problem with a two-scale expansion technique.

There are two intrinsic characteristic time scales involved. Firstly, the time \( \tau_v = nl/|q_0| \) during which the solute is transported convectively over the distance of one integral scale \( l \). Secondly, the time \( \tau_d = nl^2/d_\perp \) during which the solute is spread by local transversal dispersion over the distance of one integral scale. The ratio between the convective and the dispersive timescale defines the Péclet number

$$\text{Pe} = \frac{\tau_d}{\tau_v} = \frac{|q_0| l}{d_\perp}.$$  

(27)

In most groundwater applications the Péclet number is in the range of \( 10^2 \) – \( 10^4 \), and therefore \( \tau_d \gg \tau_v \). The long-time asymptotic behavior of the density front is reached only after large enough time compared to the (large) dispersive timescale. So the dimensionless time is introduced as 

$$\tilde{t} = \frac{t}{t_{\text{scale}}}, \quad t_{\text{scale}} = \frac{\tau_d}{\varepsilon_t},$$

(28)

where \( \varepsilon_t \ll 1 \) is a small parameter.

Finally, introducing the dimensionless velocity and density as

$$\tilde{q} = \frac{q}{|q_0|}, \quad \tilde{\rho} = \frac{\rho - \rho_+}{\rho_- - \rho_+},$$

(29)

keeping in mind that

$$\frac{\partial}{\partial x_2} = \frac{1}{l} \frac{\partial}{\partial \tilde{x}_2}, \quad \frac{\partial}{\partial x_1} = \frac{1}{l} \left( \frac{\partial}{\partial \tilde{x}_1} + \varepsilon_x \frac{\partial}{\partial \tilde{z}} \right), \quad \frac{\partial}{\partial t} = \frac{d_\perp}{nl^2} \left( \varepsilon_t \frac{\partial}{\partial \tilde{t}} \mp \text{Pe} \varepsilon_x \frac{\partial}{\partial \tilde{z}} \right),$$

(30)

and dropping the tilde notation, equations (17)-(19) and (24) are recast into

$$\varepsilon_t \frac{\partial \rho}{\partial \tilde{t}} - \frac{\partial^2 \rho}{\partial x_2^2} - d \left( \frac{\partial}{\partial x_1} + \varepsilon_x \frac{\partial}{\partial \tilde{z}} \right)^2 \rho + \text{Pe} q \cdot \nabla \rho + \text{Pe} \varepsilon_x (q_1 \pm 1) \frac{\partial \rho}{\partial \tilde{z}} = 0,$$

(31)
∇·q + ε∂q1/∂z = 0, \quad (32)

\frac{∂}{∂x_2}\left(q_1 e^{-σ\mathcal{K}} + N_g ρ\right) = \left(\frac{∂}{∂x_1} + ε\frac{∂}{∂z}\right) q_2 e^{-σ\mathcal{K}}, \quad (33)

z → ±∞ : \quad ρ = \begin{cases} 0 \\ 1 \end{cases}, \quad \langle e^{-σ\mathcal{K}}q_2 \rangle = 0, \quad \langle q_1 \rangle = ±1. \quad (34)

The sign + (−) corresponds to the case of the front moving in upward (downward) direction. In addition to the Péclet number (27), we have the dimensionless parameters

d = \frac{d∥}{d⊥}, \quad N_g = \frac{Δρk_0g}{μ|q_0|}, \quad (35)

where $N_g$ is the gravity number. The correlation function $R$ as a result of the scaling has the properties

\[ R(0) = 1, \quad \int_0^∞ R(r) \, dr = 1. \quad (36) \]

Note that $\mathcal{K}$ is a function of the fast coordinates $x$ only, whereas $ρ$ and $q$ depend on $x$, $z$, and $t$. All these functions are random processes in $x$–space. It is natural to claim that at fixed $z$ and $t$, the random fields $ρ(x)$ and $q(x)$ have the same property of translation invariancy as $\mathcal{K}$. So it is assumed that $ρ(x)$ and $q(x)$ are homogeneous random fields as well.

The term with the gravity number $N_g$ in (33) is responsible for the stabilizing gravity effect. This number ranges from zero for the tracer case, to several units for slow seepage of highly concentrated brines. Large values of $N_g$ are exceptions rather than the rule. For instance, even if $Δρ = 10^2$ kg/m$^3$, $N_g = 1$ for the typical values $k_0 = 10^{-12}$ m$^2$, and $|q_0| = 10^{-6}$ m/s. Therefore, only the cases $N_g ≪ 1$ (tracer transport) and $N_g ∼ 1$, will be considered in this paper.

### 3.1.3 Macroscale equations

The macroscale equations for the brine transport problem are derived with a traditional method, which consists of two steps. First, the macroscale solute mass balance equation is obtained. The corresponding equation has a common form and is written in terms of the average density $\langle ρ \rangle$ and the dispersive mass flux $J$, which are functions of the vertical coordinate $z$ and time $t$ only. The second step is based on the assumption of weakly heterogeneous media, i.e. small $σ$. It consists of deriving with $O(σ^2)$ accuracy the dependence of $J$ on $\langle ρ \rangle$. This rheological relation is of great interest in our analysis. In addition, a relation will be derived for the variance

\[ σ^2_ρ = \langle(ρ - \langle ρ \rangle)^2\rangle. \quad (37) \]
Obtaining the macroscale balance equation is a trivial task. It is done simply by spatial averaging of (31) in the fast coordinate space. Using the property $\langle \nabla f \rangle = 0$, valid for any homogeneous random field $f$, with the equation of incompressibility of the flow (32), we obtain

$$
\varepsilon_t \frac{\partial \langle \rho \rangle}{\partial t} + \varepsilon_x \frac{\partial J}{\partial z} = 0,
$$

(38)

$$
J = \frac{\text{Pe}}{\varepsilon_x} \langle (q_1 \mp 1)(\rho - \langle \rho \rangle) \rangle - d \frac{\partial \langle \rho \rangle}{\partial z}.
$$

(39)

Furthermore, a more convenient notation for the macroscopic balance equation is used, which we derive from (38) by differentiation:

$$
\varepsilon_t \frac{\partial \gamma}{\partial t} = \varepsilon_x \frac{\partial^2 J}{\partial z^2},
$$

(40)

$$
J = d\gamma + \text{Pe}^2 N_g \langle (q_1 \mp 1)(\rho - \langle \rho \rangle) \rangle,
$$

(41)

in which $\gamma$ denotes the dimensionless density gradient

$$
\gamma = -\varepsilon_x \text{Pe} N_g \frac{\partial \langle \rho \rangle}{\partial z}.
$$

(42)

Note that $\text{Pe} N_g = \text{Ra}$ and that $\gamma$ can be considered as a local Rayleigh number. Expression (42) is equivalent to (16) written in the original physical variables.

The averaging of (32) and (33) yields

$$
\frac{\partial}{\partial z} \langle q_1 \rangle = 0, \quad \frac{\partial}{\partial z} \langle e^{-\sigma K} q_2 \rangle = 0.
$$

(43)

The boundary conditions (34) for $q_1$ and $q_2$ remain unaffected.

The rheological relation, i.e. the expression for the dispersive flux $J$ in terms of the density gradient $\gamma$, can be found solving the so-called cell problem. The derivation of the cell problem is given in Appendix A. The solution of the cell problem is described in Appendix B. Here, only the final results are presented. The total dispersive mass flux is of the form

$$
J = d\gamma + \sigma^2 \text{Pe} I(\gamma),
$$

(44)

where the first term on the right-hand side is the microscale dispersive flux, while the second term describes macrodispersion. In the macro dispersive flux term, $I(\gamma)$ can be written as

$$
I(\gamma) = I_0(\gamma) + \varepsilon_t I_1 + \varepsilon_x^2 I_2,
$$

(45)

where the second and third term on the right-hand side are the relaxation and interaction term respectively, which are derived in Appendix B.2. Dropping
these two terms, using the smallness of $\varepsilon_t$ and $\varepsilon_x$, yields the simplest case where only the term $I_0$ is retained. For the 2-D Gaussian correlation function, in dimensionless variables

$$R(r) = e^{-\pi r^2/4},$$

the following explicit relation is obtained:

$$I(\gamma) = I_0(\gamma) = \gamma \left( 1 - \sqrt{\gamma} e^{\gamma/\pi} \text{erfc} \sqrt{\gamma/\pi} \right).$$

The dispersion coefficient is obtained by dividing the dispersive flux $J$ by the gradient $\gamma$. In the original dimensional variables, $D$ can be written as

$$D = d_\parallel + l\sigma^2|q_0|D_0(\gamma),$$

where $d_\parallel = nd_{\text{mol}} + \alpha_\parallel|q_0|$ is the local dispersion coefficient. The coefficient $D_0(\gamma) = I(\gamma)/\gamma$ multiplies the asymptotic dispersion coefficient for the tracer case, i.e. $l\sigma^2|q_0|$, and can be interpreted as a correction factor due to the stabilizing gravity effect. Table 1 presents the results for this correction factor for two types of correlation functions in two and three dimensions.

<table>
<thead>
<tr>
<th>$R(r)$</th>
<th>2-D</th>
<th>3-D</th>
</tr>
</thead>
<tbody>
<tr>
<td>$e^{-\left(\frac{r}{\ell}\right)}$</td>
<td>$\frac{1}{1-\gamma} \left( 1 - \frac{\sqrt{\gamma}}{\sqrt{1-\gamma}} \arctan \left( \frac{\sqrt{1-\gamma}}{\sqrt{\gamma}} \right) \right)$</td>
<td>$\frac{1-\gamma + \gamma \ln \gamma}{(1-\gamma)^2}$</td>
</tr>
<tr>
<td>$e^{-\pi \left(\frac{r}{\ell}\right)^2}$</td>
<td>$1 - \sqrt{\gamma} e^{\gamma/\pi} \text{erfc}(\sqrt{\gamma/\pi})$</td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Correction factor $D_0(\gamma)$ for the 2-D and 3-D exponential and Gaussian correlation functions.

Finally, we present the expression for the concentration variance

$$\sigma^2_\rho = \frac{\sigma^2_\rho}{\text{Pe}N_g^2} e^{\gamma/\pi} \frac{\gamma^{3/2}}{\text{erfc}\left(\sqrt{\gamma/\pi}\right)},$$

for the case of the 2-D Gaussian correlation function. Note that the variance in (49) is the variance of the scaled density, whereas in the numerical experiments it is defined in terms of the scaled mass fraction (referred to as concentration). As the difference between the two is at most a few percent for $\varepsilon = 0.2$, the two will be directly compared.

### 3.2 Comparison with numerical experiments

A comparison is made between the homogenization model and the numerical experiments described in Part I. We will refer to the simplest macroscopic model, i.e. with the algebraic rheological relation $I = I_0(\gamma)$, as the homogenization model. This model consists of the macroscale balance equation (40),
the dispersive flux given by (44), and the rheological relation (47). This system
is solved numerically, in order to obtain the time evolution of $\gamma(z,t)$. Subse-
quently, the density profiles are obtained by integration of $\gamma$. The system
is solved in the dimensionless variables, with parameter values chosen to match
the numerical experiments. Therefore, $l = (\sqrt{\pi}/2)\lambda = 0.886 \text{ cm}$, $\text{Pe} = l/\alpha_\parallel = 88.6$, and $d = 1$ (isotropic local dispersion). The gravity number $N_g$ depends
on the values of $q_0$ and $\Delta \rho$ in the specific experiment, according to expres-
sion (35), in which $g = 9.81 \text{ m/s}^2$, $\mu = 1 \cdot 10^{-3} \text{ kg/ms}$, and $k_0 = 5 \cdot 10^{-12} \text{ m}^2$.
In the numerical experiments, $N_g$ ranges from zero (tracer case) to 1.64.

As initial condition for $\gamma$ a smooth profile is used, instead of taking a delta-
function (corresponding to a step function in the density). The initial distribu-
tion is computed using a concentration profile from the numerical experiment
at an early time instant, corresponding to a travelled length of 10 integral
scales.

3.2.1 Concentration profiles

In Figure 8, density profiles computed with the homogenization model are
compared to the numerical experiments and to the predictions of the linear
model. Figures 8a and 8b show the cases of a relatively low and a high density
contrast, where $\varepsilon = 0.00125$ and $\varepsilon = 0.1$ respectively. The homogenization
model prediction in the latter case is very good, but shows deviations from
the experimental results for $\varepsilon = 0.00125$. However, the homogenization model
in the low-density case is still clearly better than the linear model. Note that
the predictions of the linear model, which is strictly valid only for the tracer
case, are closer to the experiments in the low-density case than in the high-
density case.

Next, we show that the homogenization model is able to predict the experi-
ments for different flow rates. Figure 9 shows the results for the largest density
difference $\varepsilon = 0.2$ and two different flow rates. In both cases, the homogeniza-
tion model generates adequate predictions of the experiments. Recall that
with the nonlinear model of Hassanizadeh and Leijnse [15], the value of the
nonlinear dispersion parameter $\beta$ has to be adapted for different flow rates.
No fitting is necessary when the homogenization model is used.

The homogenization model is based on the assumption of a small log-permea-
bility variance $\sigma^2$. Figures 8 and 9 show a satisfactory agreement between the
model predictions and the numerical experiments for $\sigma^2 = 0.1$. In Figure 10 the
model predictions are compared to the experiments for $\sigma^2 = 0.5$ and $\varepsilon = 0.2$.
Even for this large density difference there is a significant difference between
model predictions and experiments. Also plotted in Figure 10 is the best fit
with the nonlinear model of Hassanizadeh and Leijnse [15]. Though also not
Fig. 8. Density profiles predicted by the homogenization model and the linear model compared to the numerical experiment, for $\sigma^2 = 0.1$ and $q_0 = 0.6 \cdot 10^{-5} \text{ m/s}$.
(a) low-density case; (b) high-density case.

matching perfectly, this model shows a better agreement with the experiments than the homogenization model, except for the first profile plotted in Figure 10. The reason for the good match between the homogenization model and this experimental profile is that it shortly follows the profile which is taken as initial condition.
3.2.2 Concentration variances

The concentration variance is computed with (49), in which the experimental values of $\gamma$ are used. With regard to the numerical experiments, Part I gives the definitions of three different concentration variances and their relations. As discussed in Part I, the variance (or uncertainty) in the local concentration attains a maximum at the center of the brine front. In Figure 11, the maximum experimental variance is compared to the maximum theoretical variance, for density contrasts of 200 and 12.5 kg/m$^3$.

Figure 11a shows the results for $\sigma^2 = 0.1$. For $q_0 t/n\lambda > 60$ the theoretical pre-
Fig. 10. Comparison between the homogenization model, the numerical experiments, and the nonlinear model \((\beta = 4.3 \cdot 10^5)\), for \(\sigma^2 = 0.5\), \(q_0 = 0.6 \cdot 10^{-5}\) m/s, and \(\varepsilon = 0.2\).

diction is close to the experimental data for both density differences. In the short-time limit, the theoretical variance according to (49) becomes infinitely large, because \(\lim_{t \to 0} \gamma = \infty\). One of the main assumptions in the homogenization procedure is the existence of two length scales \(l\) and \(L\) that are well separated. At short times, when the transition between brine and fresh water is very sharp, the assumption \(\varepsilon_x = l/L \ll 1\) does not hold. Therefore, the homogenization theory is not able to describe the short-time behavior.

In Figure 11b the results for \(\sigma^2 = 0.5\) are plotted. For \(\sigma^2 = 0.5\), \(\sigma_{\rho}^2\) converges more rapidly to the tracer value when \(\Delta \rho\) decreases than for \(\sigma^2 = 0.1\). The experimental data plotted for \(\Delta \rho = 12.5\) in Figure 11b are almost identical to the data for the true tracer case, i.e. for \(\Delta \rho = 0\). Note the smaller difference between the experimental results for \(\Delta \rho = 200\) and for \(\Delta \rho = 12.5\) in Figure 11b compared to Figure 11a. The theoretical prediction of \(\sigma_{\rho}^2\), however, keeps increasing with decreasing \(\Delta \rho\). The homogenization theory is not able to predict the tracer case. From expression (49) it is clear that for finite \(\gamma\), \(\lim_{N_a \to 0} \sigma_{\rho}^2 = \infty\). Even for \(\varepsilon = 0.2\) the agreement between theory and experiment is much worse in Figure 11b than in Figure 11a, though the theoretical prediction still converges towards the experimental profile with time. Note also that the theoretical predictions for \(\sigma^2 = 0.5\) show more oscillations than for \(\sigma^2 = 0.1\). This is a result of the \(\gamma\) profiles being less smooth, even though a larger number of realizations was used for \(\sigma^2 = 0.5\) than for \(\sigma^2 = 0.1\). Increasing the number of realizations would yield smoother curves, but would not change their essential features.

In Figure 12, \(\sigma_{\rho}^2\) given by expression (49) is compared to the experimental \(\sigma_{\rho}^2\)
Fig. 11. Maximum concentration variance of numerical experiments compared to theoretical predictions ($q_0 = 0.6 \cdot 10^{-5}$ m/s). (a) $\sigma^2 = 0.1$; (b) $\sigma^2 = 0.5$.

at large time. For both density differences the theoretical prediction shows a satisfactory agreement with the experimental variance. The maximum value of $\sigma_c^2$ (plotted in Figure 11) is well predicted, but the theoretical profile is somewhat wider than the experimental one.
4 Homogenization model of Demidov

In contrast to [20] and Egorov, who consider averaged balance equations at the local scale, Demidov\(^1\) starts at the pore scale level. Demidov applies the homogenization method to flow around microscale heterogeneities, rather than considering heterogeneities in permeability. He considers three types of simple geometries (see Figure 13): a capillary tube, a slit, and a 2-D mesh of squares.

\(q_0 = 0.6 \cdot 10^{-3} \text{ m/s}, \sigma^2 = 0.1, \Delta \rho \text{ in kg/m}^3\).

---

\(^1\) Details of this model will be published in the PhD thesis *Upscaling of brine-transport equations in micro-heterogeneous porous media* (in Russian), by Denis Demidov, Kazan State University.
In these geometries, upward brine filtration is considered, with the flow described by the Stokes equations. The Boussinesq approximation is again adopted. Homogenization of the local equations results in the macroscale transport equation

$$\frac{\partial \langle \rho \rangle}{\partial t} + V \frac{\partial \langle \rho \rangle}{\partial z} = \frac{\partial}{\partial z} \left[ D_{tr} D_0(\gamma_l) \frac{\partial \langle \rho \rangle}{\partial z} \right],$$

(50)

where $\rho$ is again the scaled density, $V$ is the average fluid velocity, and $D_{tr}$ is the effective tracer dispersion coefficient, which is multiplied by $D_0(\gamma_l)$. Note that (50) is almost identical to the macroscopic balance equation obtained by Egorov (40). However, in Demidov’s definitions the dimensionless density gradient $\gamma_l$ is given by

$$\gamma_l = - \frac{l_p^4 g}{d_{mol} \mu} \frac{\partial \langle \rho \rangle}{\partial z},$$

(51)

where $l_p$ denotes the characteristic pore size.

For the simple cases of the capillary and the slit, explicit expressions for $D_0(\gamma_l)$ can be derived. When $l_p$ defined for the slit case is slightly adjusted compared to $l_p$ for the tube, the two solutions coincide. For the 2-D periodic arrangement of squares, the cell problem needs to be solved numerically. The solution is almost independent of $\text{Pe}_\perp$, and for $\theta = 45^\circ$ is very close to the 1-D case of the capillary tube. For $\gamma_l \to \infty$: $D_0 \sim \gamma_l^{-3/4}$. Moreover, the solution can be approximated by

$$D_0 = \frac{1 - c_1 \gamma_l + c_2 \gamma_l^4}{1 + c_3 \gamma_l^{4.75}},$$

(52)

where $c_1 = 0.0038$, $c_2 = 1.6 \cdot 10^{-9}$, and $c_3 = 4.7 \cdot 10^{-11}$.

The predictions of Demidov’s model compare well to the concentration profiles of [9] (obtained for coarse and medium sand), using one and the same ratio between the grain size and the characteristic pore size $l_p$. In order to make a comparison with the numerical experiments in macroscopically heterogeneous media, a relation between $k_0$, $l$ (or $\lambda$), and $l_p$ is needed. Note that, comparing expression (51) to (16), the factor $k_0 \lambda^2 \pi/4 (= k l^2)$ in expression (16) is substituted by $l_p^4$ in expression (51). Furthermore, the molecular diffusion coefficient $d_{mol}$ plays the role of $d_\perp$.

Consider a porous medium consisting of a bundle of capillary tubes, arranged in a 3-D lattice with periodic size $L$. For laminar (Poiseuille) flow of a Newtonian fluid through a straight circular tube with radius $l_p$, the volume flow rate is given by

$$\phi_v = -\frac{\pi l_p^4}{8\mu} \frac{\partial p}{\partial z},$$

(53)

where $p$ denotes the fluid pressure. To obtain the specific discharge, $\phi_v$ needs to be divided by the area of one periodic cell $L^2$. Expression (53) is equivalent
to Darcy’s law, when

\[ k = \frac{\pi l_p^4}{24 T L^2}, \]

(54)

where a tortuosity factor \( T \geq 1 \) is taken into account. The porosity can be expressed as

\[ n = 3\pi l_p^2 / L^2. \]

(55)

Furthermore, assume that the pore radius \( l_p \) and the heterogeneity size \( l \) are related as

\[ l_p = Sl, \]

(56)

where \( S \sim 1 \) is a scaling factor. Using expressions (55) and (56), yields

\[ \frac{l_p^4}{k l^2} = F, \]

(57)

where \( F = 24T S^2 / n \) is the factor relating \( \gamma \) to \( \gamma_l \), i.e. \( \gamma = \gamma_l / F \). For example (assuming \( n = 0.4 \)), \( F = 15 \) for \( S = 0.5 \) and \( T = 1 \), while \( F = 360 \) for \( S = 2 \) and \( T = 1.5 \). In principle, \( F \) needs to be obtained by fitting the model to the experimental results.

4.1 Comparison with numerical experiments

A comparison between Demidov’s model and the numerical experiments is made. The approximate formula (52) is used, which together with equation (50) forms a system of equations that needs to be solved numerically. In order to match the numerical experiments, the model parameters are chosen as \( D_{kr} = l \sigma^2 q_0 \), \( V = q_0 / n \), and \( \gamma_l = \gamma F \). The experiments for \( \sigma^2 = 0.1 \) can all be satisfactorily modelled using \( F = 240 \). A comparison between the computed and experimental profiles is given in Figure 14, for density differences \( \Delta \rho = 12.5 \) and 100 kg/m\(^3\). The linear model predictions are plotted as well. Figure 14 can be compared to Figure 8, which depicts Egorov’s model predictions for the same cases. For both density differences a satisfactory agreement with the numerical experiment is found.

Furthermore, Demidov’s model is able to describe the experiments for different flow rates with the same value of \( F \). Similar plots as for Egorov’s model, plotted in Figure 9, are obtained. An attempt is made to model the numerical experiments for \( \sigma^2 = 0.5 \) using the same conversion factor \( F = 240 \). Figure 15a shows that the agreement for \( \Delta \rho = 200 \) kg/m\(^3\) is reasonable, and much better than the linear model. The fit with the experiment can be improved by reducing \( F \) by a factor three. For the intermediate density difference \( \Delta \rho = 25 \) kg/m\(^3\), plotted in Figure 15, the model prediction for \( F = 240 \) is as bad as the linear model, but showing larger gradients. For \( F = 80 \), the agreement with the numerical experiment is good. In conclusion, Demidov’s
model cannot be applied to the more heterogeneous media without adjusting $F$. Note that for the tracer case, the agreement is excellent for any $F$, as the homogenization model in that case reduces to the linear model.
Fig. 15. Density predictions of Demidov’s model (using $F = 240$ and $F = 80$) compared to the numerical experiment for $\sigma^2 = 0.5$ and $q_0 = 0.6 \cdot 10^{-5}$ m/s.

4.2 Conclusions

- Demidov applies homogenization theory to flow in porous media at the pore scale level. His model can also be applied to macroscopically heterogeneous porous media, using a conversion factor $F$ for the dimensionless density gradient.
- For $\sigma^2 = 0.1$, the results of Demidov’s model are very similar to those of Egorov’s model.
For $\sigma^2 = 0.5$, the model of Demidov shows a much better agreement with the experiments than that of Egorov, thanks to the possibility of adjusting $F$.

5 Comparison and discussion

In this section, a comparison is made between the nonlinear model of Hassanizadeh [15] and the homogenization models of Egorov and Demidov. Their ability to predict the experimental data, their applicability, and their limitations are discussed. Furthermore, the macroscopic dispersive fluxes are compared. A connection between the models of Hassanizadeh and Egorov is made, based on equality of fluxes. This results in an expression for $\beta$ in terms of porous medium and flow parameters.

5.1 Comparison of predictions

In the previous sections, density profiles obtained with the different macroscopic models were compared to the results of the numerical experiments. However, the differences in the model predictions are more clearly demonstrated in the profiles of the density gradient $\gamma$. Figure 16 shows the $\gamma$ distributions at various time instants, computed with the three different models, compared to the numerical experiment. The envelopes, depicting the maxima of $\gamma$ for all times, are shown as well. Note that, to obtain the data for the figures in this section, the initial condition for the nonlinear model of Hassanizadeh [15] is taken identical to those of the homogenization models, i.e. based on an early-time experimental profile rather than a step function.

For $\Delta \rho = 100 \, \text{kg/m}^3$, depicted in Figure 16a, an excellent agreement with the experiment is found for both homogenization models. For large time, Hassanizadeh’s model with $\beta = 5 \cdot 10^5$ generates smaller gradients. A better fit can be obtained by adjusting $\beta$, but the idea behind the model is to use the same $\beta$ for every density contrast.

For the small density contrast plotted in Figure 16b, the nonlinear model of Hassanizadeh is closest to the experiments. Moreover, the model predictions of Demidov for $\Delta \rho = 6.25 \, \text{kg/m}^3$ are closer to the experiment than those of Egorov. According to Egorov’s formula for the 2-D Gaussian correlation function (47), $D_0$ approaches 1 at an infinite angle as $\gamma \downarrow 0$. In Demidov’s case, $\lim_{\gamma \to 0} (D_0) = 1 - c_1 \gamma$, so that $D_0$ approaches 1 at a finite angle. For small $\gamma$, i.e. approaching the tracer case, the approximation of Demidov is closer to reality. The nonlinear model of Hassanizadeh [15] for small density
Fig. 16. Dimensionless density gradient $\gamma$ and its envelope, predicted with the three different models in comparison with the numerical experiment for $\sigma^2 = 0.1$ and $q_0 = 0.6 \cdot 10^{-5}$ m/s.

gradients simply reduces to the classical linear model.

In Figure 17, a comparison is made with the numerical experiments for $\sigma^2 = 0.5$. As shown earlier in Figure 10, the homogenization theory of Egorov is not adequate to describe the experiments for the more heterogeneous fields. This is confirmed in Figure 17, where the computed $\gamma$ with Egorov’s model is much larger than the experimental value, regardless of the density difference. In contrast to Egorov’s model, some extent of parameter fitting is possible
Fig. 17. Dimensionless density gradient $\gamma$ and its envelope, predicted with the three different models in comparison with the numerical experiment for $\sigma^2 = 0.5$ and $q_0 = 0.6 \cdot 10^{-5}$ m/s.

with Demidov’s model. The model of Demidov shows a good agreement for $\sigma^2 = 0.5$ using $F = 80$, in both Figures 17a and b. Note that $F = 240$ is used in Figure 16 for the case $\sigma^2 = 0.1$.

The nonlinear model of Hassanizadeh [15] in Figures 17a and b shows a good agreement for both density contrasts. However, this agreement is obtained using different values for $\beta$ (see the legends in the figures). [15] and [7] claim that $\beta$ is independent of $\Delta \rho$. In other words: experiments for any density
difference can be modelled with one and the same $\beta$. Though this is the case for $\sigma^2 = 0.1$, the results for $\sigma^2 = 0.5$ contradict this statement. (See also Figure 7.)

5.2 Comparison of fluxes

The important difference between the theory of [20] and the results of the homogenization procedure [2] on one hand, and the nonlinear theory of Hassanizadeh [15] on the other hand, is the form of the dispersive flux equation. In this section, the fluxes given by the linear theory, the nonlinear theory of Hassanizadeh [15], and the homogenization/stochastic theory [20] are compared.

According to the linear Fickian theory, the dispersive mass flux in terms of the density in 1-D is given by

$$J_\rho = -D_{tr} \frac{\partial \rho}{\partial z}.$$  \hspace{1cm} (58)

In the stochastic and homogenization models, the macroscopic flux in dimensional variables is expressed as

$$J_\rho = -D_v D_0 \frac{\partial \rho}{\partial z}.$$  \hspace{1cm} (59)

It should be noted that these fluxes are different from the flux $J$, given by expression (12), in Hassanizadeh’s model. As a result of writing the salt mass balance (3) in terms of the density using the equation of state (6), the flux $J$ in mass balance equation (7) is multiplied by $\gamma_\rho$. Therefore, $J_\rho = \gamma_\rho J$.

Next, the fluxes are expressed in dimensionless form using

$$J^* = \frac{J_\rho}{J_0}, \quad J_0 = \frac{\mu d_1 D_{tr}}{k_0 g l^2}.$$  \hspace{1cm} (60)

In terms of the dimensionless density gradient $\gamma$, we obtain $J^* = \gamma$ and $J^* = I(\gamma) = D_0(\gamma)\gamma$ in the linear and homogenization/stochastic model respectively. The model of Hassanizadeh [15] yields

$$J = \gamma_\rho \frac{-1 + \sqrt{1 + 4\beta J_0 \gamma}}{2\beta J_0 \gamma}.$$  \hspace{1cm} (61)

The fluxes given by the three models are depicted in Figure 18, where for $D_{tr}$ the values of the numerical experiments for $\sigma^2 = 0.1$ and $\sigma^2 = 0.5$ are used, with $\beta = 5 \times 10^5$ and $\beta = 1 \times 10^5$ respectively. These two cases yield nearly identical fluxes according to expression (61), because (as expected) the ratio between the fitted tracer dispersivities is nearly five, yielding the same value for the
product $J_0\beta$. For the homogenization/stochastic model, $J^*$ is independent of $D_{tr}$ or $\sigma^2$. For a given density gradient $\gamma$, the dispersive mass flux for all other models is significantly lower than for the linear model. Note that the 3-D exponential correlation function used by [20] yields larger dispersive fluxes than the 2-D Gaussian.

Fig. 18. Comparison of dimensionless fluxes given by various models.

Fig. 19. Predicted $\beta$ based on expression(62) with $\beta_0 = 8 \cdot 10^{-9}/D_{tr}$, compared to the results of the numerical experiments for $\sigma^2 = 0.1$ and 0.5 ($q_0 = 0.6 \cdot 10^{-5}$ m/s).

The flux according to nonlinear relation (61) depends on $\beta$. In order to obtain
a flux equal to that in the homogenization/stochastic model, \( \beta \) should satisfy

\[
\beta = \frac{\beta_0}{D_0(\gamma)} \left( \frac{1}{D_0(\gamma)} - 1 \right),
\]

(62)

where \( \beta_0 = \gamma \rho / J_0 \) depends on medium and fluid parameters. Note that—in the absence of molecular diffusion—\( d_\perp = \alpha_\perp q_0 \) and \( D_{\text{tr}} = \alpha_{\text{tr}} q_0 \). Therefore, \( \beta \) depends on the flow rate through \( \beta_0 \sim q_0^{-2} \). The numerical experiments for \( \sigma^2 = 0.1 \) show a very similar flow rate dependence, for which a power of \( -1.85 \pm 0.13 \) is obtained. The nonlinear parameter \( \beta \) according to (62) further depends on \( \gamma \), and through \( D_0(\gamma) \) on the type of correlation function, see Table 1.

Figure 19 shows the predictions based on expression (62), for the two media with the Gaussian correlation function, using \( \beta_0 = 8 \cdot 10^{-9} / D_{\text{tr}} \). The \( \beta \) values fitted to the experimental results including the estimated error bounds (see also Figure 2) are plotted as well.

5.3 Comparison of effective dispersivities

In Part I of this paper, the macroscale dispersivities (scaled by the tracer values) of various laboratory experiments are compared against the gravity number \( N_g \). The numerical experiments for \( \sigma^2 = 0.1 \) show a similar decrease of \( A_{11}/A_{11,\text{tr}} = D_0 \) as the laboratory experiments in homogenous media, as shown in Figure 14 in Part I. The relation between \( D_0 \) and \( N_g \) is not unique, but depends on \( \sigma^2 \), see also the analysis in Appendix A. The models of [20] and Egorov and Demidov, however, suggest that \( D_0 \) is a function of the (dimensionless) density gradient rather than of the gravity number. Moreover, from the theory of Hassanizadeh [15], an apparent dispersivity can be found that is also dependent on the density gradient. When molecular diffusion is disregarded, the dispersive flux in the nonlinear model is given by equation (4), where \( D = \alpha_{\text{tr}} |q| \). Setting this flux equal to the flux given by the linear relation, i.e. Fick’s law, where \( D = \alpha_{\text{app}} |q| \), yields

\[
\frac{\alpha_{\text{app}}}{\alpha_{\text{tr}}} = \frac{2}{1 + \sqrt{1 + P \frac{\partial \rho}{\partial z}}},
\]

(63)

Note that the right-hand side of this expression also depends on \( \alpha_{\text{tr}} \), because \( P = 4 \beta \alpha_{\text{tr}} |q| \varepsilon \rho / \gamma_p \).

In the nonlinear model microscale dispersion is not treated separately, as it is in the numerical experiments and homogenization model. Therefore, in order to make a comparison with \( D_0(\gamma) \), expression (63) needs to be scaled as
\[ D_0 = \frac{D - d_\parallel}{D_{tr} - d_\parallel} = \frac{\alpha_{app} - \alpha_\parallel}{\alpha_{tr} - \alpha_\parallel} = \frac{2}{1 + \sqrt{1 + P\partial^2\rho/\partial z}} \left( \frac{\alpha_{tr}}{\alpha_{tr} - \alpha_\parallel} \right) - \frac{\alpha_\parallel}{\alpha_{tr} - \alpha_\parallel}. \]  

(64)

In Figure 20, expression (64) is compared to the solutions of [20] (for the 3-D exponential correlation function) and the homogenization theory (for the 2-D exponential and Gaussian correlation functions). See for these explicit solutions Table 1. Demidov’s results for the 2-D mesh are plotted as well, for Pe_⊥ = 90 (corresponding to the numerical experiment), where again \( F = 240 \). A comparison is made with the results of the numerical experiments. However, the experimental data are not directly comparable to the theoretical data, because no direct relation between \( D_0 \) and \( \gamma \) can be established. The dimensionless density gradient \( \gamma \) in the experiment varies in both space and time. The dispersion coefficients in Figure 20 are obtained by fitting the erfc-solution to the profiles obtained at large time (\( q_0 t/n\lambda = 100 \) and \( q_0 t/n\lambda = 80 \)). The shape of the profile at large time is determined by its travel from \( t = 0 \) up to that time. These dispersion coefficients are therefore effective time averages. The corresponding density gradients are taken from the maximum gradients of the long-time profiles. Taking the maximum gradient overestimates the average gradient in space. However, the long-time gradient underestimates the average gradient in time (although the gradient decreases only slowly in time, as can be seen for example in Figures 5–10).

Fig. 20. Predicted \( \beta \) based on expression (62) with \( \beta_0 = 8 \cdot 10^{-9}/D_{tr} \), compared to the results of the numerical experiments for \( \sigma^2 = 0.1 \) and 0.5 (\( q_0 = 0.6 \cdot 10^{-5} \) m/s).

Even though the dimensionless gradients are only roughly estimated, in Fig-
ure 20 all experimental data points for similar $\gamma$ are close. Taken at the same time but for different $\sigma^2$, the points lie practically on the same line. This line strikingly coincides with the data of Demidov. Whereas the relation between $D_0$ and $N_g$ depends on $\sigma^2$ (see Part I), the relation between $D_0$ and $\gamma$ is independent of $\sigma^2$. The data points for $\sigma^2 = 0.1$ but different times ($q_0 t/n\lambda = 80$ and 100 respectively) are close as well, indicating convergence in time.

The line depicting Hassanizadeh’s theory in Figure 20 is computed from (64), using the values $\alpha_{tr} = 0.94$ mm and $\beta = 5 \times 10^5$, fitted to the results for $\sigma^2 = 0.1$. Approximately the same line is obtained for $\sigma^2 = 0.5$, where $\alpha_{tr} = 4.5$ mm and $\beta = 1 \times 10^5$, again indicating no dependence on $\sigma^2$. A unique relation between $D_0$ and $\gamma$ suggests that the product $\beta \alpha_{tr}$ is constant. This is consistent with the fact that $\beta$ decreases with $\sigma^2$, as depicted in Figure 2.

Though the experimental data are not directly comparable to the theoretical relations for $D_0$, the data points in Figure 20 lie within the area enclosed by the theoretical curves. A good agreement is obtained with Demidov’s data for a 2-D periodic medium. For large $\gamma$ the experimental data are close to the theoretical predictions for the 2-D Gaussian and exponential correlation functions. Comparing the solution for the exponential correlation function in three [20] and in two dimensions (Egorov), shows that the density effect for a given $\gamma$ is stronger in the 2-D case. The effect is also stronger for the Gaussian correlation function than for the exponential. In all cases however, the dispersion coefficient decreases to 10% of its tracer value when $\gamma$ is in the order of 10. Demidov’s results for large $\gamma$ are close to the results for both 2-D correlation functions. For small $\gamma$, however, Demidov’s result coincides with the result of Hassanizadeh’s model. The latter reduces to the linear model for infinitely small $\gamma$. Therefore, Demidov’s model yields a more realistic description than Egorov’s model when small density contrasts are considered.

6 Summary

The second order term in Hassanizadeh’s nonlinear equation for the dispersive mass flux involves an additional dispersion parameter $\beta$. [15] claim that experiments for any density contrast can be modelled using the same $\beta$ for a specific medium and flow rate. Their findings are confirmed by [7], who analyzes the experiments of [6] in homogeneous media. The model of Hassanizadeh [15] is developed for essentially homogeneous media. In the present study, the model predictions are compared to the results of a series of numerical experiments in weakly heterogeneous media, i.e. media characterized by a small correlation scale $\lambda$ and a small log-permeability variance $\sigma^2$. Indeed, we find that for $\sigma^2 = 0.1$, even though the best-fit $\beta$ value is variable, the experiments for all densities can be satisfactorily modelled with the same $\beta$. Furthermore, the
findings for $\sigma^2 = 0.1$ confirm the flow-rate dependence of $\beta$ that is reported by [7] and by [9].

In this study, the travel-time dependency of $\beta$ is studied as well. The best-fit value of $\beta$ increases slowly with time, and does not reach an asymptotic value within the time-range of the numerical experiments. Moreover, for media with stronger heterogeneities ($\sigma^2 = 0.5$) it is no longer possible to describe the experiments for different density contrasts using the same $\beta$. When $\Delta \rho$ decreases, the value of $\beta$ needs to be reduced considerably in order to match the experimental results. The non-uniqueness of $\beta$ implies that the value of $\beta$ is not known beforehand, but needs to be fitted to the specific experiment.

The linear model, which is strictly only valid for tracer transport, is still often used for density- and/or viscosity-dependent dispersion problems. Then, the dispersivity is adjusted in order to match the experimental data, in contradiction with the basic premise of the classical dispersion theory, which considers dispersivity to be a medium property. This argument against using the linear dispersion model also holds for the nonlinear model, which still involves a fitting procedure (and is more complex). The advantage of the nonlinear model, in particular for small $\sigma^2$, is that $\beta$ needs to be determined less often. Moreover, with two parameters a better fit is achieved than with the linear model.

Another thing that should be noted is that in the derivation of the theory of Hassanizadeh [15], stabilizing gravity effects are not explicitly accounted for, while these are the main cause of the nonlinear effect. The theory of [20] and the homogenization theory [2] are both derived from the physical processes at the local scale. These processes include rotational gravity-flow induced by horizontal density gradients as a result of local velocity variations. Upscaling of these local processes results in a system of macroscale equations in which the macroscopic dispersion coefficient depends on the mean density gradient.

The approach of [20] or the homogenization approach can be applied to different types of media, i.e. two- or three-dimensional with different log-permeability correlation functions, see the results in Table 1. An important limitation of these approaches, however, is the assumption of a uni-directional mean flow (in vertical direction). The homogenization model of Egorov is based on the same assumptions as the numerical experiments and does not require any fitting, or additional parameters. The model predictions show a satisfactory to good agreement with the numerical experiments for $\sigma^2 = 0.1$. Unfortunately, the theory is only applicable to weakly heterogeneous media, i.e. $\sigma^2 \ll 1$. For $\sigma^2 = 0.5$ the homogenization model cannot adequately describe the numerical experiments.

Another nice result of the homogenization procedure is the expression for the
concentration variance. For \( \sigma^2 = 0.1, N_g \geq 0.2, \) and large enough time, a satisfactory agreement is found between the predicted and experimental concentration variances. At short times or for small \( N_g, \) however, the theoretical variance becomes infinitely large.

Demidov applies the homogenization method to microscale heterogeneous media. The different pore geometries (capillary tube, slit, and 2-D mesh of squares) with an appropriate choice of the characteristic pore sizes yield a very similar decrease in the effective dispersion coefficient. The macroscale transport equation is identical to that in Welty and Gelhar and Egorov’s models. Using a factor which relates the characteristic pore size to the permeability and correlation length, Demidov’s model can also describe the experiments in weakly heterogeneous media. The agreement with the numerical experiments for \( \sigma^2 = 0.1 \) is good. However, for \( \sigma^2 = 0.5 \) the scaling parameter needs to be adjusted in order to obtain a satisfactory agreement. In a way this can be seen as an advantage of the model. Using the same value for the scaling parameter, all experiments for \( \sigma^2 = 0.5 \) can be modelled, whereas the model of Egorov cannot be applied in this case at all.

The theory of [20] as well as the homogenization models suggest that the scaled macroscale dispersion coefficient (or dispersivity) \( D_0(\gamma) \) is a function of the (dimensionless) density gradient rather than of the gravity number \( N_g. \)

The results of the numerical experiments support this conclusion. Whereas the relation between \( D_0 \) and \( N_g \) still depends on \( \sigma^2, \) the dependence on \( \gamma \) is unique for a certain type of correlation function. Moreover, for the theory of Hassanizadeh [15] an apparent dispersivity can be derived which depends on \( \gamma. \) For this relationship to be unique, the product \( \beta \alpha_{tr} \) needs to be constant. In addition, a relation between \( \beta \) and \( \gamma \) is derived based on equality of fluxes in the two different formulations.

7 Conclusions

Hassanizadeh and Leijnse’s nonlinear equation for the dispersive mass flux contains an additional term in comparison to linear Fick’s law. This second order term involves an extra dispersion parameter \( \beta. \) Hassanizadeh and Leijnse [15] claim that experiments for any density contrast can be modelled using the same \( \beta \) for a specific medium and flow rate. Their findings are confirmed by Schotting et al. [7], who analyzes the experiments of Moser [6] in homogeneous media. The model of Hassanizadeh and Leijnse [15] is developed for essentially homogeneous media. In the present study, the model predictions are compared to the results of a series of numerical experiments in weakly heterogeneous media, i.e. media characterized by a small correlation scale \( \lambda \) and a small log-permeability variance \( \sigma^2. \) Indeed, we find that for \( \sigma^2 = 0.1, \)
even though the best-fit $\beta$ value is variable, the experiments for all densities can be satisfactorily modelled with the same $\beta$. Furthermore, the findings for $\sigma^2 = 0.1$ confirm the flow-rate dependence of $\beta$ that is reported by Schotting et al. [7] and by Watson et al. [9].

In this paper, the travel-time dependency of $\beta$ is studied as well. The best-fit value of $\beta$ increases slowly with time, and does not reach an asymptotic value within the time-range of the numerical experiments. Moreover, for media with stronger heterogeneities, i.e. $\sigma^2 = 0.5$, it is no longer possible to describe the experiments for different $\Delta \rho$ using the same $\beta$. When $\Delta \rho$ decreases, the value of $\beta$ needs to be reduced considerably in order to match the experimental results. The non-uniqueness of $\beta$, i.e. its dependence on medium characteristics, flow rate, travel time/distance, and possibly on the density difference, is the main disadvantage of the nonlinear model. It implies that the value of $\beta$ is not known a priori, but needs to be fitted to the specific experiment. In this sense the nonlinear model has no advantage over the linear model. The linear model, which is strictly only valid for tracer transport, is still often used for density- and/or viscosity-dependent dispersion problems. Then, the dispersivity $\alpha$ is adjusted in order to match the experimental data, in contradiction with the statement that dispersivity is a medium parameter. This argument against using the linear model also holds for the $\beta$ parameter. Using the nonlinear model still involves a fitting procedure, and is more complex. However, with two parameters a better fit is achieved than the optimal fit with the linear model.

Another thing to note about the theory of Hassanizadeh and Leijnse [15] is that in their derivation stabilizing gravity effects are not explicitly accounted for, while these are the main cause of the nonlinear effect. The theory of Welty and Gelhar and the homogenization theory are both derived from the physical processes at the local scale. These processes include rotational gravity-flow induced by horizontal density gradients due to local velocity variations. Upscaling of these local processes results in a system of macroscale equations, in which the macroscopic dispersion coefficient depends on the mean density gradient.

The approach of Welty and Gelhar or the homogenization approach can be applied to different types of media, i.e. two- or three-dimensional with different log-permeability correlation functions, see Table 1. The homogenization model is based on the same assumptions as the numerical experiments and does not require any fitting, or additional parameters. The model predictions show a satisfactory agreement with the numerical experiments for $\sigma^2 = 0.1$. Unfortunately, the theory is only applicable to weakly heterogeneous media, i.e. $\sigma^2 \ll 1$. For $\sigma^2 = 0.5$ the homogenization model can not adequately describe the numerical experiments.
Another nice result of the homogenization procedure is the expression for the concentration variance \( \sigma_c^2 \). For \( \sigma^2 = 0.1 \), \( N_g \geq 0.2 \), and large enough time \( (q_0 t/n \lambda \geq 60) \), a satisfactory agreement is found between the predicted and experimental concentration variance. At short times or for small \( N_g \), however, the theoretical variance becomes infinitely large.

The theories of Hassanizadeh and Leijnse \([15]\), of Welty and Gelhar \([20]\), and the homogenization model all suggest that the scaled macroscale dispersion coefficient (or dispersivity) \( D_0(\gamma) \) (or \( \alpha/\alpha_{tr} \)) is a function of the (dimensionless) density gradient rather than of the gravity number \( N_g \). The results of the numerical experiments support this conclusion. Whereas the relation between \( \alpha/\alpha_{tr} \) and \( N_g \) still depends on \( \sigma^2 \), the dependence on \( \gamma \) is unique for a certain type of correlation function.

**Appendix**

**A Derivation of the cell problem**

The solution of the system (31)-(33) is decomposed into a series of powers of the small parameter \( \sigma \):

\[
\rho = \rho^{(0)}(z, t) + \sigma \rho^{(1)}(z, t; x) + O(\sigma^2),
\]

\[
q_1 = \pm 1 + \sigma q_1^{(1)}(z, t; x) + O(\sigma^2),
\]

\[
q_2 = 0 + \sigma q_2^{(1)}(z, t; x) + O(\sigma^2).
\]

Additionally, it is required that for \( n \geq 1 \)

\[
\langle \rho^{(n)} \rangle = 0, \quad \langle q_1^{(n)} \rangle = 0, \quad \left\langle \sum_{k=1}^{n} \frac{(-1)^{(n-k)} q_2^{(k)} K^{n-k}}{(n-k)!} \right\rangle = 0.
\]

The first condition leads to the equality of \( \rho^{(0)} \) to the mean density \( \langle \rho \rangle \), which appears in the macroscale balance equation. The other two conditions follow from the boundary condition for the vertical velocity component and the condition for the mean transverse pressure gradient, see (34).

After substitution of the series (A.1)-(A.3) into (32) and (33), the terms having equal powers of \( \sigma \) are gathered. For \( n = 1 \) we obtain

\[
\frac{\partial q_1^{(1)}}{\partial x_1} + \frac{\partial q_2^{(1)}}{\partial x_2} + \varepsilon_x \frac{\partial q_1^{(1)}}{\partial z} = 0,
\]

\[
\frac{\partial}{\partial x_2} \left( q_1^{(1)} K + N_g \rho^{(1)} \right) = \left( \frac{\partial}{\partial x_1} + \varepsilon_x \frac{\partial}{\partial z} \right) q_2^{(1)}.
\]
Substituting the series into (31) yields

\[ A_0 + \sigma A_1 + \sigma^2 A_2 + \cdots = 0, \]  

(A.7)

where (taking into account (A.5))

\[ A_0 = \varepsilon_t \frac{\partial \rho^{(0)}}{\partial t} - d\varepsilon_x^2 \frac{\partial^2 \rho^{(0)}}{\partial z^2}, \]  

(A.8)

\[ A_1 = \varepsilon_t \frac{\partial \rho^{(1)}}{\partial t} - \frac{\partial^2 \rho^{(1)}}{\partial x^2} - d \left( \frac{\partial}{\partial x_1} + \varepsilon_x \frac{\partial}{\partial z} \right)^2 \rho^{(1)} \pm Pe \frac{\partial \rho^{(1)}}{\partial x_1} + Pe \varepsilon_x q_1^{(1)} \frac{\partial \rho^{(0)}}{\partial z}. \]  

(A.9)

We require \( A_n \) to be independent of the fast coordinates, i.e. \( A_n = \langle A_n \rangle \). With this requirement, relation (A.7) becomes an alternative expression for the macroscopic balance equation (40). The first of these requirements, \( A_0 = \langle A_0 \rangle \), is automatically satisfied, while the second, \( A_1 = \langle A_1 \rangle = 0 \), together with (A.5) and (A.6) constitutes a system of coupled linear equations for deriving \( \rho^{(1)} \), \( q_1^{(1)} \), and \( q_2^{(1)} \). These three equations in the terminology of homogenization theory are referred to as the cell problem. It is important to note that the macroscopic density gradient \( \frac{\partial \rho^{(0)}}{\partial z} \) is a parameter of the cell problem. After solution of the cell problem, the macroscopic dispersive flux \( J \) and uncertainty \( \sigma_\rho^2 \) are calculated using

\[ J = d\gamma + \sigma^2 Pe^2 \cdot F \rho^{(1)} q_1^{(1)} \rho^{(1)} \],  

(A.10)

\[ \sigma_\rho^2 = \sigma^2 \rho^{(1)} q_1^{(1)} \rho^{(1)} \],  

(A.11)

which are obtained by substitution of the series (A.1) and (A.2) in (37) and (41). The parameter \( \gamma \) (42) will appear in these relations.

The same cell problem at \( \varepsilon_x = 0 \) was derived by Welty and Gelhar [20] for the case of a three-dimensional exponential correlation function. Welty and Gelhar’s analysis was based on Fourier transformation of the cell problem. The same transformation is applied here, and after elimination of \( q_2^{(1)} \) the cell problem is rewritten in the frequency plane as

\[ k_2^2(v + c - 1) = \left( \varepsilon_x \frac{\partial}{\partial z} + ik_1 \right)^2 v, \]  

(A.12)

\[ \varepsilon_t \frac{\partial c}{\partial t} \pm iPe k_1 c - \gamma v + k_2^2 c = d \left( \varepsilon_x \frac{\partial}{\partial z} + ik_1 \right)^2 c, \]  

(A.13)

where

\[ c = \mp N_x \frac{F \rho^{(1)}}{FK}, \quad v = \mp \frac{F q_1^{(1)}}{FK}. \]  

(A.14)
and \( \mathcal{F} \) denotes the Fourier transform

\[
\mathcal{F} f = \int_{-\infty}^{+\infty} e^{ik \cdot x} f(x) dx_1 dx_2.
\]

(A.15)

Using a standard technique, see Gelhar [25], the macroscopic flux \( J \) can be expressed in terms of \( c, v \), and the Fourier transform \( \phi(|k|) = \mathcal{F} R \) of the correlation function:

\[
J = d\gamma + \sigma^2 Pe \cdot I,
\]

(A.16)

\[
I = \frac{Pe}{(2\pi)^2} \int_{-\infty}^{+\infty} \phi(|k|) \text{Re}(c^*v) dk_1 dk_2,
\]

(A.17)

where the asterisk denotes the complex conjugate. A similar expression is obtained for the uncertainty

\[
\sigma_ho^2 = \sigma^2 Pe^{-1} N_g^{-2} I_\sigma,
\]

(A.18)

\[
I_\sigma = \frac{Pe}{(2\pi)^2} \int_{-\infty}^{+\infty} \phi(|k|) |c|^2 dk_1 dk_2.
\]

(A.19)

The first term on the right hand side of (A.16) is the microscale dispersive flux, while the second term is the macroscale dispersive flux. In order to obtain the rheological relations for \( I \) and \( I_\sigma \) in terms of the density gradient \( \gamma \), the system (A.12)-(A.13) needs to be solved. The solutions \( c \) and \( v \) are used to calculate the integrals (A.17) and (A.19). In the general case this procedure defines a nonlinear, nonlocal in space \( z \) and time \( t \) rheological operator \( \gamma \rightarrow I \), and an analogous operator \( \gamma \rightarrow I_\sigma \).

B Solution of the cell problem

To simplify the derivation of the rheological operator \( I \), the smallness of the parameters \( \varepsilon_x \) and \( \varepsilon_t \) is used. The simplest algebraic rheological relationship can be derived when the terms with \( \varepsilon_x \) and \( \varepsilon_t \) in equations (A.12) and (A.13) are disregarded.

B.1 The algebraic rheological relation

Posing \( \varepsilon_x = \varepsilon_t = 0 \) reduces equations (A.12) and (A.13) to the algebraic system

\[
v + c = 1, \quad \pm Pe k_1 c + k_2^2 c - \gamma v = 0.
\]

(B.1)
From (B.1) we observe that the large term \( Pe k_1 c \) dominates the other terms in (B.1). The only way to arrive at a nontrivial solution is by re-normalization of the parameter \( k_1 \) with a large factor. Without loss of generality, this factor is chosen to be \( Pe \), i.e.

\[
\tilde{k}_1 = Pe k_1. \tag{B.2}
\]

The necessity of the re-normalization is a common conclusion, see e.g. Gelhar [25], and Welty and Gelhar [20]. It means that the major contribution to the integrals (A.17) and (A.19) is made up of a small neighborhood around \( k_1 = 0 \). As a result of the re-normalization, we obtain

\[
c = c_0(\gamma) = \frac{\gamma}{\kappa + \gamma}, \quad v = v_0(\gamma) = \frac{\kappa}{\kappa + \gamma}, \quad \kappa = k_2^2 \pm i\tilde{k}_1, \tag{B.3}
\]

and rewrite the integrals (A.17) and (A.19) as

\[
I = \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} \phi(|k_2|) \text{Re}(c^* v) \, d\tilde{k}_1 dk_2, \tag{B.4}
\]

\[
I_\sigma = \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} \phi(|k_2|) |c|^2 \, d\tilde{k}_1 dk_2. \tag{B.5}
\]

These integrals are very similar to (A.17) and (A.19), but, as a result of the re-normalization, \( \phi \) becomes a function of \(|k_2|\) instead of \(|k|\). This allows us to take the inner integrals in (B.4) and (B.5) explicitly and find \( I \) and \( I_\sigma \) as

\[
I = I_0(\gamma) = \frac{\gamma}{4\pi} \int_{-\infty}^{+\infty} \phi(|k_2|) \frac{k_2^2}{\gamma + k_2^2} \, dk_2, \tag{B.6}
\]

\[
I_\sigma = \frac{\gamma^2}{4\pi} \int_{-\infty}^{+\infty} \phi(|k_2|) \frac{1}{\gamma + k_2^2} \, dk_2. \tag{B.7}
\]

If the spectrum \( \phi \) is specified, the derivations of the functions \( I_0(\gamma) \) and \( I_\sigma \) are simple exercises in calculus. For the two-dimensional isotropic Gaussian correlation function, with spectrum \( \phi(k) = 4e^{-k^2/\pi} \), we obtain

\[
I_0(\gamma) = \gamma \left( 1 - \sqrt{\gamma} e^{\gamma/\pi} \text{erfc} \sqrt{\gamma/\pi} \right), \tag{B.8}
\]

\[
I_\sigma(\gamma) = \gamma^{3/2} e^{\gamma/\pi} \text{erfc} \sqrt{\gamma/\pi}. \tag{B.9}
\]

These results apply to the case of the numerical experiment. For the case of a three-dimensional exponential correlation function, Welty and Gelhar [20] derived an analogous expression, see Table 1.

Note that the asymptotic behavior of \( I_0 \) at small \( \gamma \) can be expressed universally as

\[
\gamma \to 0 : \quad I_0 = \gamma - a_0 \gamma^{3/2} + O(\gamma^{3/2}), \tag{B.10}
\]

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where the constant \( a \) depends on the integral characteristic of the correlation function:

\[
a = \frac{\phi(0)}{4} = \frac{\pi}{2} \int_{0}^{\infty} R(r) r \, dr.
\]  

(B.11)

The limit of small \( \gamma \) corresponds to the classical tracer case. In this case the rheological relation is reduced to the well known result \( I = \gamma \). In the original dimensional coordinates, this implies a linear dependence of the dispersive flux on the density gradient, with a constant dispersion coefficient \( \sigma^2 l \), see Dagan [24] or Gelhar [25]. This classical relation is valid for large travel time, consistent with \( \varepsilon_x = \varepsilon_t = 0 \).

The macroscale balance equation (40), expression (A.16), and rheological relation \( I = I_0(\gamma) \), form a system of equations for deriving the average density gradient \( \gamma \) and the dispersive mass flux \( J \). This algebraic macroscale model is used for comparison with the numerical experiments.

**B.2 Including derivatives in the cell problem**

In Section B.1, the simplest case of the algebraic cell problem, i.e. for \( \varepsilon_x = \varepsilon_t = 0 \) was discussed. Let us now take into account the terms that have been disregarded so far. Again, the normalizing parameter \( \tilde{k}_1 \) is used in the analysis. Furthermore, we suggest that \( \text{Pe} \varepsilon_x \gg 1 \) and use the smallness of \( \text{Pe}^{-1} \) to replace the operators

\[
\left( \varepsilon_x \frac{\partial}{\partial z} + i \text{Pe}^{-1} \tilde{k}_1 \right)^2
\]

on the right hand sides of (A.12) and (A.13) with \( \varepsilon_x^2 \frac{\partial^2}{\partial z^2} \). As a result, we obtain

\[
k_2^2(v + c - 1) = \varepsilon_x^2 \frac{\partial^2 v}{\partial z^2},
\]

\[
\varepsilon_t \frac{\partial c}{\partial t} \mp i \tilde{k}_1 c - \gamma v + k_2^2 c = d \varepsilon_x^2 \frac{\partial^2 c}{\partial z^2}.
\]

(B.12)

(B.13)

If not only the leading terms \( c_0 \) and \( v_0 \), but also the next orders are retained, i.e.

\[
c = c_0(\gamma) + \varepsilon_t c_1 + \varepsilon_x^2 c_2,
\]

\[
v = v_0(\gamma) + \varepsilon_t v_1 + \varepsilon_x^2 v_2,
\]

(B.14)

(B.15)
we obtain the independent problems

\begin{align}
  v_1 + c_1 &= 0, \quad \kappa c_1 - \gamma v_1 = -\frac{\partial c_0}{\partial t}, \\
  k_2^2 (v_2 + c_2) &= \frac{\partial^2 v_0}{\partial z^2}, \quad \kappa c_2 - \gamma v_2 = \frac{\partial^2 c_0}{\partial z^2},
\end{align}

(B.16) (B.17)

for determination of \(v_1, c_1 \) and \(v_2, c_2\) respectively. After solving these problems, the rheological operator \(I\) can be expressed as

\[ I = I_0(\gamma) + \varepsilon t I_1 + \varepsilon^2 I_2, \]

(B.18)

where

\[ I_n = \frac{1}{(2\pi)^2} \int_{-\infty}^{+\infty} \phi(|k_2|) \text{Re} (c_0^* v_n + v_0^* c_n) \, d\tilde{k}_1 \, dk_2, \quad n = 1, 2. \]

(B.19)

We will refer to the first corrective term in (B.18) as the ”relaxation” term, and to the second as the ”interaction” term.

It is obvious from substitution \(\tilde{k}_1 \rightarrow -\tilde{k}_1\) that the integrals \(I_n\) are independent on the sign \(\pm\) in the definition of \(\kappa = k_2^2 \pm i\tilde{k}_1\), indicating the flow direction (up or down, respectively). Therefore, to simplify the presentation, the + sign is used instead of \(\pm\), so that

\[ \kappa = k_2^2 + i\tilde{k}_1 \]

(B.20)

B.2.1 The relaxation term

The solution of the cell problem (B.16) is given by

\[ c_1 = -v_1 = \frac{-1}{\kappa + \gamma} \frac{\partial c_0(\gamma)}{\partial t} = -\frac{\kappa}{(\kappa + \gamma)^3} \frac{\partial \gamma}{\partial t}. \]

(B.21)

To determine \(I_1\), the complex \(c_0^* v_1 + v_0^* c_1\) in (B.19) is rewritten as \(c_1 - 2c_0^* c_1\). Both inner integrals in (B.19), i.e.

\[ \int_{-\infty}^{\infty} c_1 \, d\tilde{k}_1 \quad \text{and} \quad \int_{-\infty}^{\infty} c_0^* c_1 \, d\tilde{k}_1 \]

are calculated.
can be taken explicitly, exploiting the Residue Theorem. For the first integral we obtain
\[
\int_{-\infty}^{\infty} c_1 d\tilde{k}_1 = -\frac{\partial\gamma}{\partial t} \int_{-\infty}^{\infty} \frac{\kappa d\tilde{k}_1}{(\kappa + \gamma)^3} =
\]
\[
= i \frac{\partial\gamma}{\partial t} \int_{-\infty}^{i\infty} \frac{\kappa d\kappa}{(\kappa + \gamma)^3} = -2\pi i \frac{\partial\gamma}{\partial t} \text{Res} \left( \frac{\kappa}{(\kappa + \gamma)^3}, \gamma \right) = 0. \quad (B.22)
\]
Along the same line
\[
-\left( \frac{\partial\gamma}{\partial t} \right)^{-1} \int_{-\infty}^{\infty} c_0^* c_1 d\tilde{k}_1 = \int_{-\infty}^{\infty} \frac{\kappa \gamma d\tilde{k}_1}{(\kappa + \gamma)^3 (\gamma + \kappa^*)} =
\]
\[
= \int_{-\infty}^{\infty} \frac{\kappa \gamma d\tilde{k}_1}{(\kappa + \gamma)^3 (\gamma + 2k_2^2 - \kappa)} = \frac{1}{i} \int_{-\infty}^{i\infty} \frac{\gamma d\kappa}{(\kappa + \gamma)^3 (\gamma + 2k_2^2 - \kappa)} =
\]
\[
= 2\pi \gamma \text{Res} \left( \frac{\kappa}{(\kappa + \gamma)^3 (\gamma + 2k_2^2 - \kappa)}, \gamma + 2k_2^2 \right) = \frac{\pi \gamma (\gamma + 2k_2^2)}{4(\gamma + k_2^2)^3}. \quad (B.23)
\]
As a result, the relaxation term has the form
\[
I_1 = F_1(\gamma) \frac{\partial\gamma}{\partial t}, \quad (B.24)
\]
\[
F_1(\gamma) = \frac{\gamma}{4\pi} \int_0^\infty \phi(k_2) \frac{\gamma + 2k_2^2}{(\gamma + k_2^2)^3} dk_2. \quad (B.25)
\]
Again, this integral can be explicitly derived if the spectrum \( \phi \) is specified. For the 2-D isotropic Gaussian correlation function we obtain
\[
F_1(\gamma) = \frac{5}{8\pi} + \frac{\gamma}{4\pi^2} + \frac{e^{\gamma/\pi} \text{erfc} \sqrt{\gamma/\pi}}{16\sqrt{\pi}} \left( 5 - \frac{12\gamma}{\pi} - \frac{4\gamma^2}{\pi^2} \right). \quad (B.26)
\]

B.2.2 The interaction term

The solution of the cell problem (B.17) is given by
\[
c_2 = \frac{dk_2^2 - \gamma}{k_2^2(\kappa + \gamma)} \frac{\partial^2 c_0}{\partial z^2}, \quad v_2 = -\frac{dk_2^2 + \kappa}{k_2^2(\kappa + \gamma)} \frac{\partial^2 c_0}{\partial z^2}. \quad (B.27)
\]
Taking into account that
\[
c_0^* = 1 - v_0^* = \frac{\gamma}{\kappa^* + \gamma} = \frac{\gamma}{\gamma + 2k_2^2 - \kappa}, \quad (B.28)
\]
we obtain
\[ c_0^* v_2 + v_0^* c_2 = \left( \frac{d}{\kappa + \gamma} - \frac{2(d+1)\gamma}{(\kappa + \gamma)(\gamma + 2k_2^2 - \kappa)} \right) \frac{\partial^2 c_0}{\partial z^2}. \] (B.29)

Using that
\[ \frac{\partial^2 c_0}{\partial z^2} = \frac{\kappa}{(\kappa + \gamma)^2} \frac{\partial^2 \gamma}{\partial z^2} - \frac{2\kappa}{(\kappa + \gamma)^3} \left( \frac{\partial \gamma}{\partial z} \right)^2, \] (B.30)
and exploiting the Residue Theorem in the same manner as for the relaxation term, we find the interaction term in the form
\[ I_2 = -(d+1) \left( F_1(\gamma) \frac{\partial^2 \gamma}{\partial z^2} + F_2(\gamma) \left( \frac{\partial \gamma}{\partial z} \right)^2 \right), \] (B.31)
where \( F_1(\gamma) \) is defined by (B.25), and \( F_2 \) as
\[ F_2(\gamma) = -\frac{\gamma}{4\pi} \int_0^{\infty} \phi(k_2) \frac{\gamma + 2k_2^2}{(\gamma + k_2^2)^4} dk_2. \] (B.32)

For the 2-D isotropic Gaussian correlation function this results in
\[ F_2(\gamma) = -\frac{7}{16\pi\gamma} + \frac{1}{3\pi^2} + \frac{\gamma}{12\pi^3} + \frac{\mathrm{e}^{\gamma/\pi}}{96\gamma\sqrt{\gamma}} \left( -21 + \frac{30\gamma}{\pi} - \frac{36\gamma^2}{\pi^2} - \frac{8\gamma^3}{\pi^3} \right). \] (B.33)

B.2.3 The relaxation-interaction macroscopic model

The functions \( F_1(\gamma) \), \( F_2(\gamma) \) and expressions (B.24), (B.31) define rheological relation (B.18). This relation together with the macroscopic balance equation (40) forms a macroscopic model of the brine transport process under consideration. This relaxation-interaction macroscale model can be simplified introducing a new pair of functions \( A(\gamma) \) and \( U(\gamma) \), related to \( F_1(\gamma) \) and \( F_2(\gamma) \) by
\[ \frac{d}{d\gamma} \left( \frac{F_1}{A} \right) = \frac{F_2}{A}, \quad \frac{dU}{d\gamma} = \frac{F_1}{A}. \] (B.34)

In terms of these new functions, rheological relation (B.18) is written as
\[ I = I_0(\gamma) + A(\gamma) \left( \varepsilon_t \frac{\partial U}{\partial t} - \varepsilon_x^2 (d+1) \frac{\partial^2 U}{\partial z^2} \right), \] (B.35)
References


