

A macroscopic two-length-scale model for natural convection in porous media driven by a species-concentration gradient

Stefan Gasow¹, Andrey V. Kuznetsov², Marc Avila^{1,3}, and Yan Jin^{1,3*}

¹ *Center of Applied Space Technology and Microgravity (ZARM), University of Bremen, 28359, Bremen, Germany*

² *Department of Mechanical and Aerospace Engineering, North Carolina State University, Raleigh, North Carolina 27695-7910, USA*

³ *MAPEX Center for Materials and Processes, University of Bremen, 28359, Bremen, Germany*

* *Corresponding author: Yan Jin (yan.jin@zarm.uni-bremen.de)*

Abstract

The modeling of natural convection in porous media is receiving increased interest due to its significance in environmental and engineering problems. State-of-the-art simulations are based on the classic macroscopic Darcy-Oberbeck-Boussinesq (DOB) equations, which are widely accepted to capture the underlying physics of convection in porous media provided the Darcy number, Da , is small. In this paper we analyze and extend the recent pore-resolved DNS of Gasow *et al.* (2020) and show that the macroscopic diffusion, which is neglected in DOB, is of the same order (with respect to Da) as the buoyancy force and the Darcy drag. Consequently, the macroscopic diffusion must be modelled even if the value of Da is small. We propose a “two-length-scale diffusion” (TLSD) model, in which the effect of the pore scale on the momentum transport is approximated with a macroscopic diffusion term. This term is determined by both the macroscopic length scale and the pore scale. It includes a transport coefficient that solely depends on the pore-scale geometry. Simulations of our model render a more accurate Sherwood number, root-mean-square (r.m.s.) of the mass concentration, and r.m.s. of the velocity than simulations that employ the DOB equations. In particular, we find that the Sherwood number Sh increases with decreasing porosity and with increasing Schmidt number (Sc). In addition, for high values of Ra and high porosities, Sh scales nonlinearly. These trends agree with the DNS, but are not captured in the DOB simulations.

Key words: convection in porous media, boundary layer stability, buoyant boundary layers, macroscopic modeling

1. Introduction

The realization of long-term storage of CO₂ in deep saline aquifers (Orr 2009; Basbug & Gumrah 2009; Pamukcu & Gumrah 2009; Huppert & Neufeld 2014; Metz *et al.* 2005; Michael *et al.* 2009), the provision of large scale thermal-energy storage systems (Heyde & Schmitz 2017; Singh 2010), and the increase of efficiency of geothermal energy extraction (Ghoreishi-Madiseh *et al.* 2013; Böttcher *et al.* 2016) are examples of emerging engineering technologies that have the potential to slow down climate change. Natural convection in porous media is a fundamental process relevant to these applications (Hewitt *et al.* 2012; Liang *et al.* 2018; Wen *et al.* 2018a; Liu *et al.* 2020a; Hewitt 2020). In general, it describes the flow of fluid in a saturated porous medium between two infinite horizontal plates driven by a temperature or species concentration difference. The variation of temperature or species concentration results in the variation of the density, which induces the buoyancy force.

In this paper, we focus on the natural convection in porous media driven by a species concentration gradient. Compared with convective heat transfer, convective mass transfer is usually characterized by high Schmidt numbers (Sc) and unlike thermal-energy, the mass cannot penetrate the surfaces of solid obstacles. In the absence of a porous medium, the natural convective fluid flow is governed by the dimensionless Rayleigh number, which describes the buoyancy-to-diffusion ratio (Kunes 2012). In the presence of a porous medium, a Rayleigh-Darcy number (hereafter Rayleigh number, Ra) is introduced; it is a modification of the conventional Rayleigh number, which takes the effect of the porous matrix into account (Nield 1994). Mass transfer in natural convection is characterized by the Sherwood number (Sh), which is the ratio of the total mass transfer rate (by convection and mass diffusion) to the diffusive mass transfer rate. The onset of natural convection occurs when Sh exceeds unity. Sh quantifies the efficiency of the mass transfer enhancement due to natural convection.

Besides field research studies (Arts *et al.* 2008) and laboratory experiments (Kneafsey & Pruess 2010; Faisal *et al.* 2015), numerical simulation is another established tool for understanding convection in porous media. Two approaches are available for the simulation of convection in porous media: pore-scale resolving direct numerical simulations (DNS) and macroscopic (volume-averaged) simulations. Macroscopic simulations are widely employed in modeling convection in porous media (Nield & Bejan 2017), due to their significantly lower computational costs. The first macroscopic model for fluid flow in porous media was proposed by Darcy (1856). Whitaker (1969) proposed the most commonly used macroscopic equations for the conservation of volume-averaged quantities. Using Whitaker's approach, the Darcy-

Oberbeck-Boussinesq (DOB) equations can be derived, as shown in Nield & Bejan (2017). This set of equations has been often used in recent studies, see Hewitt *et al.* (2012, 2013, 2014), Wen *et al.* (2015), Paoli *et al.* (2016), and Pirozzoli *et al.* (2021) as examples. A deficiency of the DOB equations is the underlying assumption that convection in porous media is uniquely determined by the Rayleigh number, in which the pore scale is combined with the macroscopic length scale. This simplification could however be at the root of reported discrepancies between numerical simulations and experiments. For example, most numerical studies based on the DOB equations indicate a linear scaling of Sh versus Ra in the ultimate regime ($Ra \geq 5,000$), whereas the experiments by Neufeld *et al.* (2010) and Keene & Goldstein (2015) exhibited a nonlinear scaling. The experiments by Backhaus *et al.* (2011) in a Hele-Shaw cell, where the flow obeys the Darcy law but there is no porous matrix, also exhibited a nonlinear scaling. However, recent studies showed that nonlinear scaling observed in Hele-Shaw experiments may be related to the 3- dimensionality of the flow (Letelier *et al.* 2019; De Paoli *et al.* 2020). In a recent study of 3-dimensional DOB simulation, Pirozzoli *et al.* (2021) indicated that the nonlinear scaling can occur in 3-dimensional flows at very high Rayleigh numbers. This could be related to supercells at the boundary, which are the footprint of megaplumes dominating the interior part of the flow.

Another possible reason for the nonlinear scaling is related to non-Darcy effects induced by the porous matrix. Various studies have been performed to analyze non-Darcy effects in natural convection in porous media. For example, Shao *et al.* (2016) and Wang & Tan (2009) included the Brinkman term (which is a Laplacian term that is included to model the effect of macroscopic velocity gradients on the momentum transport) in their simulations of convection at low Ra numbers ($Ra \leq 5,000$). However, the study of Vasseur *et al.* (1989) concluded that the Brinkman term is significant only for large Darcy numbers. Mijic *et al.* (2014) and Das *et al.* (2016) included the Forchheimer term in their models to account for the effect of turbulence. In recent years, increasing attention is paid to hydrodynamic dispersion in porous media, see Hidalgo & Carrera (2009), Yang & Vafai (2011), Ghesmat *et al.* (2011), MacMinn *et al.* (2012), Wang *et al.* (2016), Liang *et al.* (2018), Wen *et al.* (2018b), Fahs *et al.* (2020), Jouybari *et al.* (2020) and Liu *et al.* (2020b). It is sometimes also referred to as thermal dispersion for heat transfer problems (Pedras & de Lemos 2008), or mass dispersion for mass transfer problems (Mesquita & de Lemos 2004). A Fickian dispersion tensor introduced by Bear (1961) is often used to model the hydrodynamic dispersion. These studies show that hydrodynamic dispersion can have significant effects on convection in porous media, at least for high Darcy number problems. Gelhar *et al.* (1992), Lallemand-Barres & Peaudecerf (1978), Neuman (1990), and

Liang *et al.* (2018) indicated that the hydrodynamic dispersion is also important at low Darcy numbers, since dispersion at the macro-scale (macrodispersivity) is dependent on the scale of the system, rather than the grain size. In a recent study, however, Zech *et al.* (2019) showed that dispersion at the macro-scale varied widely and did not show any clear effect on the scale of solute plumes.

In the DNS, the Navier-Stokes equations coupled to a convection-diffusion equation for the species concentration (or temperature for heat transfer) are solved, whereby the smallest scale of the porous matrix is resolved. Due to the high computational costs, this approach has so far only been used for simple geometries of porous matrices (Minkowycz *et al.* 2006; Torabi *et al.* 2017). Although DNS is too expensive for engineering applications, it is a powerful tool to gain a better understanding of the physics of convection in porous media and serves as a foundation for developing macroscopic models. Recently, we performed pore-scale-resolving DNS of natural convection in porous media composed of a simple porous matrix (Gasow *et al.* 2020). Our DNS results showed that the boundary layer thickness for convection in porous media is determined by the pore size instead of the Rayleigh number. This is distinctly different from classical DOB simulations (Huppert & Neufeld 2014). We also showed that the scaling for the Sherwood number depends on the porosity and the pore-scale parameters and observed that the scaling law becomes nonlinear for porous media with sufficiently high porosity. Furthermore, the computed flow patterns exhibited motions with large length scales, close to the size of the whole domain, which were not found in DOB simulations. In another recent numerical study, Liu *et al.* (2020a) observed that the Nusselt number increases with a decrease in the porosity, while the Rayleigh-Darcy number is kept constant. This trend cannot be captured by the DOB equations. Liu *et al.* (2020a) also indicated that the ratio of the pore scale to the thickness of the thermal boundary layer has a significant effect on the scaling of the Nusselt number versus Ra . A scaling crossover occurs when the thickness of the thermal boundary is comparable to the pore scale. Therefore, the discrepancy between the DOB solutions and the experiments could arise due to pore-scale effects.

In this paper, we develop a new macroscopic model for natural convection in porous media, which accounts for pore-scale effects. Our model is based on a detailed analysis of the DNS simulations of Gasow *et al.* (2020) and additional DNS carried out here. The model involves a coefficient, which depends solely on the pore-scale geometry. This coefficient must be determined a priori. For each pore-scale geometry, this coefficient is determined with a single DNS performed with a fixed set of parameters. Subsequently, we show that the simulations of

the model agree with our DNS results (e.g. results with respect to the Sherwood number, mean species concentration, r.m.s. species concentration, and velocity) in wide ranges of pore size, Rayleigh, Schmidt, and Darcy numbers.

2. Governing equations and numerical methods

We consider natural convection in a porous medium domain bounded by two walls (figure 1), which is the porous equivalent to the classical Raleigh-Bénard cell (Hewitt 2020). The computational domain is two-dimensional, and it has a width-to-height ratio $L/H = 2$. Two different geometries of the generic porous matrix are studied. They are composed of aligned (figure 1(b)) or staggered (figure 1(c)) square obstacles. The analysis in this study is mainly based on the results of the first porous matrix, while the sensitivity of our model coefficient to the pore scale geometry is examined with the second porous matrix. In both cases, the periodically arranged square obstacles with the size d are a distance s apart in the horizontal and vertical directions. The geometry of a REV of the simulated porous medium is a square with a side length s , containing one obstacle.

Constant species concentrations, c_1 and c_0 , are maintained at the upper and lower walls of the domain, respectively. The difference of the species concentrations at the upper and lower walls leads to density differences, which drives natural convection in the domain. The horizontal boundary conditions are periodic, whereas the no-slip boundary condition is used at the upper and lower walls and on the surfaces of the obstacles. And because mass cannot penetrate the solid matrix of the porous medium, no mass transfer is assumed at the interface, hence homogeneous Neumann boundary conditions are used at the obstacles for the species concentration. Similar setups have been adopted in other numerical studies of convection in porous media, see Javaheri *et al.* (2010), Hewitt *et al.* (2012), Wen *et al.* (2019), and Hewitt (2020) as examples.

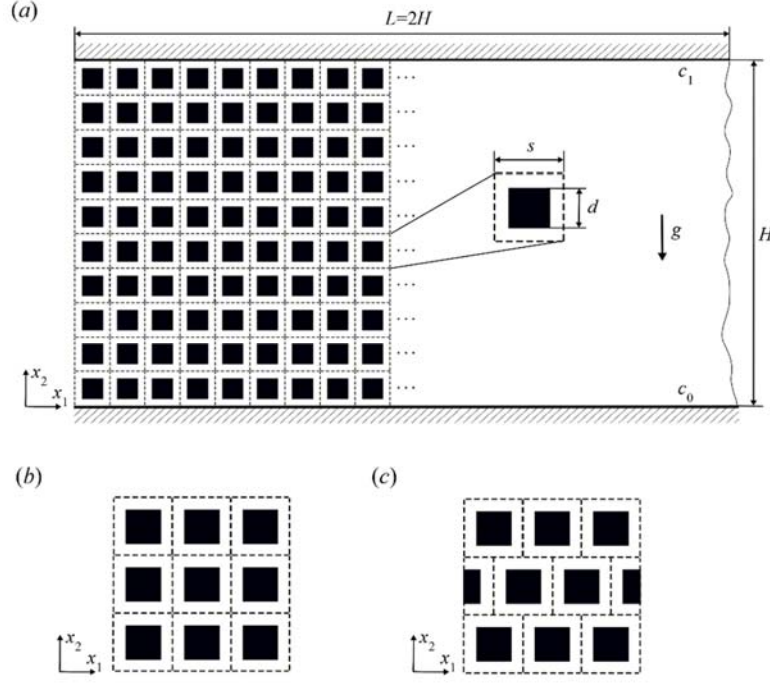


Figure 1. Structure of the computational domain occupied by a regular porous matrix, with a magnified view of a single REV, used for the DNS (a). A constant species concentration difference at the top and bottom walls and periodic boundary conditions in the horizontal direction are utilized. The porous matrix inside the domain is composed of aligned (b) or staggered square obstacles (c).

2.1 Governing equations for DNS

DNS studies were performed to gain insights into the physics of natural convection in the porous medium, to determine the coefficients for the macroscopic model, as well as to obtain the validation data. The governing equations for DNS of natural convection in porous media are the Navier-Stokes equations and the species transport equation. In the flow field, the local species concentration differences are small; hence, the Boussinesq approximation is used to account for the buoyancy force (Herwig 2013). Using Einstein's summation convention, the governing microscopic equations for natural convection in porous media are as follows:

$$\frac{\partial u_i}{\partial x_i} = 0, \quad (1)$$

$$\frac{\partial u_i}{\partial t} + \frac{\partial(u_i u_j)}{\partial x_j} = -\frac{\partial p}{\partial x_i} + \nu \frac{\partial^2 u_i}{\partial x_j^2} + \beta g_i (c - c_0), \quad (2)$$

$$\frac{\partial c}{\partial t} + \frac{\partial(u_i c)}{\partial x_i} = D_f \frac{\partial^2 c}{\partial x_j^2}, \quad (3)$$

where ν , D_f , u_i , p , g_i , and c are the kinematic viscosity, the mass diffusivity, the i th component of the velocity vector, the pressure, the i th component of the gravity vector, and the species concentration, respectively. The concentration expansion coefficient is defined as $\beta = \beta(c_0) = -1/\rho_0 (\partial\rho/\partial c)_{c_0}$, see Herwig & Moschalski (2009), where ρ is the fluid density.

The Sherwood number Sh is calculated from the DNS as the ratio of the total mass transfer rate $\dot{m}$ (by convection and diffusion) to the mass transfer $\dot{m}_{diff}$ (by diffusion only) across the lower or upper wall (Baehr & Stephan 2006):

$$Sh = \dot{m}/\dot{m}_{diff} = \frac{\int_w \overline{\frac{\partial c}{\partial c_2}} dA}{\int_w \left. \overline{\frac{\partial c}{\partial c_2}} \right|_{Ra_f=0} dA}, \quad (4)$$

where the sign $\bar{}$ denotes the time averaging operator, while the subscript w denotes either the upper or lower wall surface.

2.2 Macroscopic equations

The macroscopic equations are obtained by averaging the Navier-Stokes equations and the species transport equation (1)-(3) over each REV (see figure 1). This method of averaging is similar to the one used in de Lemos (2012), however, de Lemos (2012) carried out a time and volume averaging over each respective REV, while we performed only volume averaging. The macroscopic equations read:

$$\frac{\partial \tilde{u}_i}{\partial \tilde{x}_i} = 0, \quad (5)$$

$$\frac{\partial \tilde{u}_i}{\partial \tilde{t}} + \frac{\partial (\tilde{u}_i \tilde{u}_i / \phi)}{\partial \tilde{x}_j} + \frac{\partial (\phi \langle \tilde{u}_i \tilde{u}_j \rangle^i)}{\partial \tilde{x}_j} = - \frac{\partial (\phi \langle p \rangle^i)}{\partial \tilde{x}_i} + \nu \frac{\partial^2 \tilde{u}_i}{\partial \tilde{x}_j^2} - \phi \beta g_i (\tilde{c} - c_0) - \phi \tilde{R}_i, \quad (6)$$

$$\frac{\partial (\phi \tilde{c})}{\partial \tilde{t}} + \frac{\partial (\tilde{u}_i \tilde{c})}{\partial \tilde{x}_i} + \frac{\partial (\phi \langle \tilde{u}_i \tilde{c} \rangle^i)}{\partial \tilde{x}_j} = D_m \frac{\partial^2 \tilde{c}}{\partial \tilde{x}_j^2}, \quad (7)$$

where the sign $\tilde{}$ denotes a REV-volume-averaged quantity. The operator $\langle \rangle^i$ denotes the intrinsic volume-averaging in the fluid phase, which is adopted from Whitaker (1986). The left superscript i denotes the intrinsic deviation of a volume-averaged quantity, e.g. $\tilde{u}_i = u_i - \langle u_i \rangle^i$. The porosity ϕ is defined as $\phi = V_{void-space}/V_{total}$, $\tilde{u}_i = \phi \langle u_i \rangle^i$ is the volume-averaged velocity, which is often referred to as the superficial velocity, and $\tilde{c} = \langle c \rangle^i$ is the intrinsic averaged mass concentration. The subscript m denotes an effective property in the

volume-averaged equations, e.g., D_m is the effective mass diffusivity. Simulations of small domains are needed to determine the value of D_m for a specific pore-scale geometry (see Gasow *et al.* 2020).

The terms $\phi\langle^i u_i u_j\rangle^i$, $\phi\langle^i u_i c\rangle^i$, and $\check{R}_i$ are the momentum dispersion, mass dispersion, and total drag, respectively. The momentum and mass dispersion terms have been neglected in our model due to the underlying assumptions for convection in porous media with low Darcy numbers (see Appendix A1). Since the local Reynolds number $Re_K = |\check{\mathbf{u}}|\sqrt{K}/\nu$ in our simulations is generally smaller than unity (Gasow *et al.* 2020), the Forchheimer term in $\check{R}_i$ can be also neglected (Nield & Bejan 2017). The effects of the macroscopic velocity gradient on $\check{R}_i$ can be modelled with a Laplacian term, which was first proposed by Brinkman (1949) and then was extensively studied and improved, see Rao *et al.* (2020), Zaripov *et al.* (2019), Zhao *et al.* (2018), Liu *et al.* (2007), Valdes-Parada *et al.* (2007), Vafai (2005), Starov & Zhdanov (2001), and Ochoa-Tapia & Whitaker (1995) as examples. Here, we model the sum of the total drag $\check{R}_i$ and the diffusion term $\nu(\partial^2 \check{u}_i / \partial \check{x}_j^2)$ in equation (6) as

$$\nu \frac{\partial^2 \check{u}_i}{\partial \check{x}_j^2} + \check{R}_i = \frac{\nu}{K} \check{u}_i + \nu_m \frac{\partial^2 \check{u}_i}{\partial \check{x}_j^2}, \quad (8)$$

where K and ν_m are the permeability and effective viscosity of the porous medium. Simulations of small domains are needed to determine their values a priori (see Gasow *et al.* (2020) for details of how they were determined). The macroscopic momentum equation (6) is hence simplified to

$$\frac{\partial \check{u}_i}{\partial \check{t}} + \frac{\partial(\check{u}_i \check{u}_i / \phi)}{\partial \check{x}_j} = - \frac{\partial(\phi \langle p \rangle^i)}{\partial \check{x}_i} + \nu_m \frac{\partial^2 \check{u}_i}{\partial \check{x}_j^2} - \phi \beta g_i (\check{c} - c_0) - \phi \frac{\nu}{K} \check{u}_i. \quad (9)$$

2.3 Two-length-scale diffusion assumption

Normalizing the governing equations (5), (9) and (7) using the characteristic concentration difference $\Delta c = c_1 - c_0$, velocity $u_m = \beta \Delta c g K / \nu$, length H , and time $t_m = H / u_m$, the following dimensionless macroscopic equations are obtained

$$\frac{\partial \hat{u}_i}{\partial \hat{x}_i} = 0, \quad (10)$$

$$\frac{\partial \hat{u}_i}{\partial \hat{t}} + \frac{\partial(\hat{u}_i \hat{u}_i / \phi)}{\partial \hat{x}_j} = - \frac{\partial(\phi \langle \hat{p} \rangle^i)}{\partial \hat{x}_i} + \frac{a_v Sc}{\gamma_m Ra} \frac{\partial^2 \hat{u}_i}{\partial \hat{x}_j^2} - \frac{\phi Sc}{\gamma_m Ra Da} z_i \hat{c} - \frac{\phi Sc}{\gamma_m Ra Da} \hat{u}_i, \quad (11)$$

$$\frac{\partial(\phi\hat{c})}{\partial\hat{t}} + \frac{\partial(\hat{u}_i\hat{c})}{\partial\hat{x}_i} = \frac{1}{Ra} \frac{\partial^2\hat{c}}{\partial\hat{x}_j^2}, \quad (12)$$

where $\hat{\cdot}$ denotes a dimensionless volume-averaged quantity, $\hat{c}$ is the dimensionless volume averaged species concentration defined as $\hat{c} = (\langle c \rangle^i - c_0)/(c_1 - c_0)$, $a_v = \nu_m/\nu$ is the ratio of the effective viscosity ν_m to the molecular viscosity of the fluid ν , and $\gamma_m = D_m/D_f$ is the ratio of the effective mass diffusivity D_m to the mass diffusivity of the fluid D_f . The Rayleigh number in equations (11) and (12) is defined by using the common definition of this parameter for natural convection in porous media, as in Nield (1994):

$$Ra \equiv \frac{Ra_f Da}{\gamma_m} = \frac{H\beta\Delta c g K}{D_m \nu}. \quad (13)$$

The Schmidt number is defined as

$$Sc = \frac{\nu}{D_f}. \quad (14)$$

The Darcy number is defined as

$$Da = \frac{K}{H^2}. \quad (15)$$

By assuming that a_v is independent of Da and taking the leading order terms with respect to $1/Da$ in equation (11), one obtains the well-known DOB equations. However, we reported in our recent DNS study (Gasow *et al.* 2020) that the boundary layer thickness is determined by the pore size, which is characterized by $\sqrt{K}$. In addition, similar profiles for temporally and horizontally averaged quantities are observed when the distance from the wall is normalized with the pore size. Therefore, the Laplacian term $(a_v Sc/\gamma_m Ra)(\partial^2 \hat{u}_i/\partial \hat{x}_j^2)$ in equation (11) is expected to scale as $1/K$ and should be of order $1/Da$, exactly as the Darcy term $-(\phi Sc/\gamma_m Ra Da)\hat{u}_i$ and the buoyancy force term $-(\phi Sc/\gamma_m Ra Da)z_i \hat{c}$. Hence, the Laplacian term in the macroscopic equation cannot be neglected even if the Darcy number is small.

We here propose a model for the effective viscosity ν_m based on a two-length-scale diffusion (TLSD) hypothesis, in which the macroscopic diffusion is determined by the pore size, characterized by $\sqrt{K}$, and the distance between the lower and upper boundaries H . Our TLSD hypothesis is supported by our recent DNS (Gasow *et al.* 2020), where we showed that natural convection in porous media is determined by these two length scales. The pore size characterizes the boundary layer thickness and the size of proto-plumes, whereas the distance between the two walls determines the size of mega-plumes.

Based on the TLSL assumption stated above, a_v is modeled as:

$$a_v = \frac{\nu_m}{\nu} = \frac{a_v^*}{Da}, \quad (16)$$

where a_v^* is a constant assumed to be solely determined by the pore-scale geometry of the porous matrix. Note that the two length scales $\sqrt{K}$ and H are combined in Da . At the upper and lower walls, we imposed constant species concentrations, c_1 and c_0 , respectively, and the no-slip boundary condition.

It should be noted that only the leading order terms of Da for diffusion are kept in the macroscopic equations (10)-(12). As $Da \rightarrow 0$, the macroscopic governing equations can be further simplified to

$$\frac{\partial \hat{u}_i}{\partial \hat{x}_i} = 0, \quad (17)$$

$$\frac{\partial \hat{p}}{\partial \hat{x}_i} + \hat{c} z_i + \hat{u}_i = \frac{a_v^*}{\phi} \frac{\partial^2 \hat{u}_i}{\partial \hat{x}_j^2}, \quad (18)$$

$$\frac{\partial \hat{c}}{\partial \hat{t}} + \frac{\partial (\hat{u}_i \hat{c})}{\partial \hat{x}_i} = \frac{1}{Ra} \frac{\partial^2 \hat{c}}{\partial \hat{x}_j^2}, \quad (19)$$

where $\hat{p} = RaDa\langle\tilde{p}\rangle^i/\gamma_m Sc$ is the normalized pressure. The dimensionless time is modified to be $\hat{t} = \tilde{t}/\phi$. These macroscopic equations (17)-(19) become the DOB equations if a_v^* is set to zero. When the DOB equations are solved, only the velocity component in the wall-normal direction is set to zero at the upper and lower walls. This boundary condition was also used in other DOB simulations, see Hewitt *et al.* (2014) and Wen *et al.* (2015) as examples. In this paper, the macroscopic simulations were carried out by solving equations (10)-(12), so that the effect of the Darcy number can be assessed. The Sherwood number for the macroscopic model simulations is defined using the same definition as for the DNS, which is given in equation (4).

2.4 Numerical method

For the simulations, a finite volume-method (FVM) was utilized. The solvers were developed by using the open source code package OpenFoam 6. The spatial discretization was implemented by a second-order central-difference scheme. For time derivatives, the second-order implicit backward method was used. For the correction and coupling of the pressure and velocity fields, the Pressure-Implicit scheme with Splitting of Operators (PISO) algorithm was used (Versteeg & Malalasekera 2007). A stabilized preconditioned (bi-)conjugate gradient

solver was utilized to solve the pressure field and the momentum and species concentration equations. We have performed the code validation for our DNS solver extensively in our previous studies (Jin *et al.* 2015, 2017; Uth *et al.* 2016; Gasow *et al.* 2020).

3. Studied test cases

3.1. Description of the test cases

We continued selected DNS cases of Gasow *et al.* (2020) to improve the statistics and thus to allow a more thorough validation of our hypothesis. In addition, we also computed these cases by solving the macroscopic equations (10)-(12) with our two-length-scale diffusion model. The Rayleigh numbers Ra are up to 20,000 and the Schmidt numbers Sc are 1 and 250. The ranges of geometrical parameters of the studied test cases are given in table 1. For both DNS and macroscopic simulation cases $u_i = 0$ and $c = (c_1 - c_0)/2$ were used as initial conditions.

s/d	1.175 – 5
ϕ	0.28 – 0.96
γ_m	0.16 – 0.92
a_v^*	$2.4 \times 10^{-9} - 5.18 \times 10^{-5}$
H/s	10 – 100
Da	$7 \times 10^{-8} - 1.7 \times 10^{-4}$

Table 1. Range of geometrical parameters for the studied test cases.

To obtain representative statistical results, the time averaging, denoted by the sign $\bar{}$, of the respective variable was performed after the flow and mass concentration fields reached a statistically steady state. As an example, the time evolutions of the instantaneous Sherwood number for the DNS case with $H/s = 100$, $s/d = 1.5$, $Ra = 20,000$, and $Sc = 250$ are shown in figure 2. The time averaging of the Sherwood number has been started after the time marked by the red dashed line. At least 200 dimensionless time units H/u_m are calculated to obtain the statistical results.

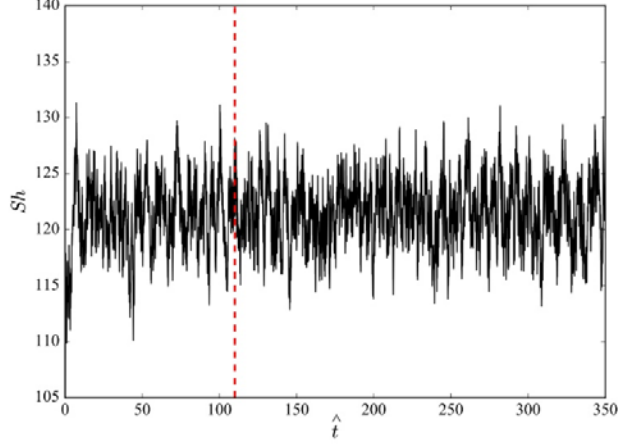


Figure 2. The time evolution of the instantaneous Sherwood number for the DNS case with $H/s = 50$; $s/d = 1.5$; $Ra = 20,000$; and $Sc = 250$. The dashed red line marks the time at which the time-averaging is started. $\hat{t} = tu_m/H$ is the dimensionless time.

3.2. Determination of the model coefficient

The coefficient a_v^* for a specific pore-scale geometry cannot be computed a priori with simulations of small domains (as for the other model parameters). Here, we empirically determine a_v^* by simulating natural convection within the specific pore-scale geometry with fixed values of H/s , Sc , and Ra . Since we only keep the leading order terms of the order Da in our model equations, a test case with a sufficiently small Darcy number should be used to ensure that the higher order terms of Da are negligible. In particular, we performed a parametric study for $a_v^*(\phi)$ while keeping $H/s = 20$, $Sc = 250$, and $Ra = 20,000$ fixed. These parameter values were selected because the Sherwood number from DNS marginally changes as H/s is increased (i.e. as the pore size is decreased), hence the effect of higher order terms of Da on Sh can be safely neglected. The value of a_v^* is selected for each considered porosity value, so that the Sherwood number from the macroscopic simulation matches the DNS results. Figure 3 shows the dependence of a_v^* on the porosity ϕ .

We expect that a_v^* is a geometrical parameter that is independent of Sc , Ra , and Da . This will be examined later in section 5. The value of a_v^* only mildly changes when the porous matrix is switched from aligned squares to staggered squares. According to our DNS results, a_v^* can be well correlated by

$$a_v^*(\phi) = 7.5 \times 10^{-5} \phi^9. \quad (20)$$

This correlation has reasonable accuracy for $0.28 < \phi < 0.95$. However, variations of pore-scale geometries used in this study are limited. In particular, the flow structures in randomly

packed porous matrices may be distinctly different from those in regularly packed porous matrices (Liu *et al.* 2020). Studies with more pore-scale geometries are needed to test the generality of equation (20).

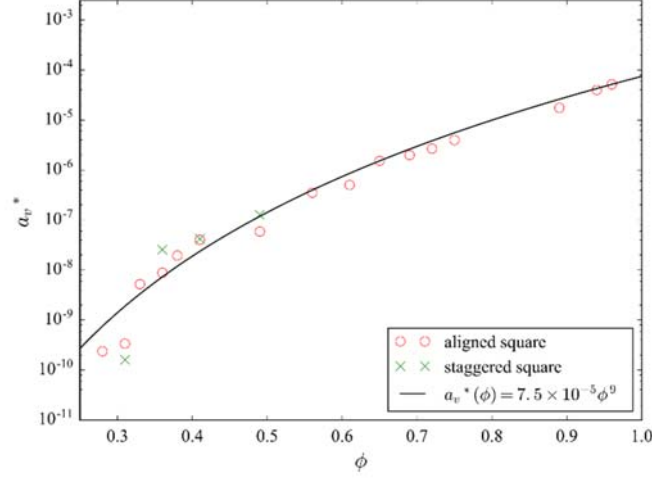


Figure 3. Dependence of the model coefficient a_v^* on the porosity ϕ , for porous matrices composed of aligned and staggered square obstacles.

3.3. Mesh and time step independence study

The mesh and time step independence study for the DNS cases have already been performed in our previous work, see Gasow *et al.* (2020). Here we focus on the influence of the mesh and time step on the macroscopic simulation results (solution of equations (10)-(12)). The numerical results for the Sherwood number are shown in table 2. At least 200 dimensionless time units H/u_m were calculated to obtain the statistical results.

ID	Mesh resolution ($N_x \times N_y$)	Mesh size	Co_{max}	Sh
a	800×1200	960 000	0.2	105.3
b	1000×1600	1 600 000	0.2	110.1
c	2000×3000	6 400 000	0.2	120.1
d	3200×2200	7 040 000	0.2	122
e	2000×3200	6 400 000	0.6	122.4

f	<u>2000×3200</u>	<u>6 400 000</u>	<u>0.8</u>	<u>121.5</u>
g	2000×3200	6 400 000	1.1	115.8

Table 2. Influence of mesh and time step on the Sherwood number Sh . The test case with $H/s = 50$, $s/d = 1.5$, $Ra = 20,000$, and $Sc = 250$ is used in the parametric study. The cases ‘c’, ‘d’, ‘e’, and ‘f’ are considered to be mesh and time step independent. The mesh resolution and maximum Courant number of the case ‘f’ (underlined) are used in all cases of macroscopic simulation.

The results of the resolution study show that the Sherwood number is under-predicted if the mesh resolution is too low (see table 2 cases ‘a’ and ‘b’) or the maximum Courant number is too high (see table 2 case ‘g’). According to the mesh/time step independence study, $Co_{max} = 0.8$ and mesh resolution 2000×3200 (case ‘f’) were used for all cases of macroscopic simulation. The numerical error of Sh in the macroscopic simulations is estimated to be 2.8 %, which is the maximum variation of Sh in the cases ‘c’, ‘d’, ‘e’, and ‘f’. All simulations were performed on the clusters of the HLRN (North-German Supercomputing Alliance), using 2x Intel Cascade Lake Platinum 9242 CPUs (CLX-AP) with 96 cores per node. The DNS cases use up to 7.2×10^7 mesh cells, which requires a parallel computing time of 1,200 hours using 384 processors. The macroscopic simulation cases use up to 6.4×10^6 mesh cells.

4. DNS results

In this section, we focus on an a priori verification of the TLSD hypothesis. The model results are compared with the DNS results in section 5.

4.1. Budget of the macroscopic kinetic energy

The budget for the time- and line-averaged macroscopic kinetic energy $\langle \bar{K} \rangle^{x_1} = \frac{1}{2} \langle \tilde{u}_i^2 \rangle^{x_1}$ was calculated from the DNS for $Ra = 20,000$, $s/d = 1.5$, $s/d = 1.25$, $Sc = 250$ and Da in the range $3.5 \times 10^{-7} - 3.5 \times 10^{-5}$. By averaging the momentum equation (2) over REV, taking the dot-product with the superficial velocity $\tilde{u}_i = \phi \langle u_i \rangle^i$, and then averaging in time- and in the horizontal direction x_1 , we obtained the following equation for $\langle \bar{K} \rangle^{x_1}$

$$-\overline{\langle \tilde{u}_i \phi \left(\frac{\partial(u_i u_j)}{\partial x_j} \right)^i \rangle^{x_1}} - \overline{\langle \tilde{u}_i \phi \left(\frac{\partial p}{\partial x_i} \right)^i \rangle^{x_1}} + \overline{\langle \tilde{u}_i \phi \left(\nu \frac{\partial^2 u_i}{\partial x_j^2} \right)^i \rangle^{x_1}} + \overline{\langle \tilde{u}_i \phi \langle \beta g_i (c - c_0) \rangle^i \rangle^{x_1}} = 0. \quad (21)$$

Equation (21) shows that the budget for $\langle \bar{K} \rangle^{x_1}$ includes

- the production by the buoyancy force, $K_{buoy} = \overline{\langle \tilde{u}_i \phi \langle \beta g_i (c - c_0) \rangle^i \rangle^{x_1}}$;

- the loss due to viscous dissipation, $K_{diff} = \overline{\langle \tilde{u}_i \phi \langle v \frac{\partial^2 u_i}{\partial x_j^2} \rangle^i \rangle^{x1}}$;
- the loss due to pressure gradient, $K_{pres} = -\overline{\langle \tilde{u}_i \phi \langle \frac{\partial p}{\partial x_i} \rangle^i \rangle^{x1}}$;
- the transport due to convection, $K_{conv} = -\overline{\langle \tilde{u}_i \phi \langle \frac{\partial (u_i u_j)}{\partial x_j} \rangle^i \rangle^{x1}}$.

In the DOB equations, the Darcy term (Darcy drag) is the only source of losses of macroscopic kinetic energy. The Darcy losses read

$$K_{Darcy} = -\phi(v/K)\langle \tilde{u}_i^2 \rangle^{x1}. \quad (22)$$

The budget of $\langle \bar{K} \rangle^{x1}$ is studied using the test case with $s/d = 1.5$ ($\phi = 0.56$), $H/s = 20$, $Ra = 20,000$, $Da = 8.8 \times 10^{-6}$, and $Sc = 250$. Figure 4 shows the distribution of K_{buoy} , K_{diff} , K_{pres} , and K_{conv} in the wall-normal direction. They are normalized with the characteristic kinetic energy $K_{mean} = 1/2 u_m^2$ or K_{buoy} . The distance from the lower wall is normalized with the pore size s . It is evident that more macroscopic kinetic energy is produced by the buoyancy force in the central region than in the region close to the wall. The transport of $\langle \bar{K} \rangle^{x1}$ due to convection is much smaller than K_{buoy} , so it can be neglected. $-K_{diff}$ and $-K_{pres}$ are the losses of the macroscopic kinetic energy. Both $-K_{diff}$ and $-K_{pres}$ increase with increasing distance from the wall x_2/s . The loss of $\langle \bar{K} \rangle^{x1}$ in the region close to the wall is mainly due to the pressure gradient $-K_{pres}$.

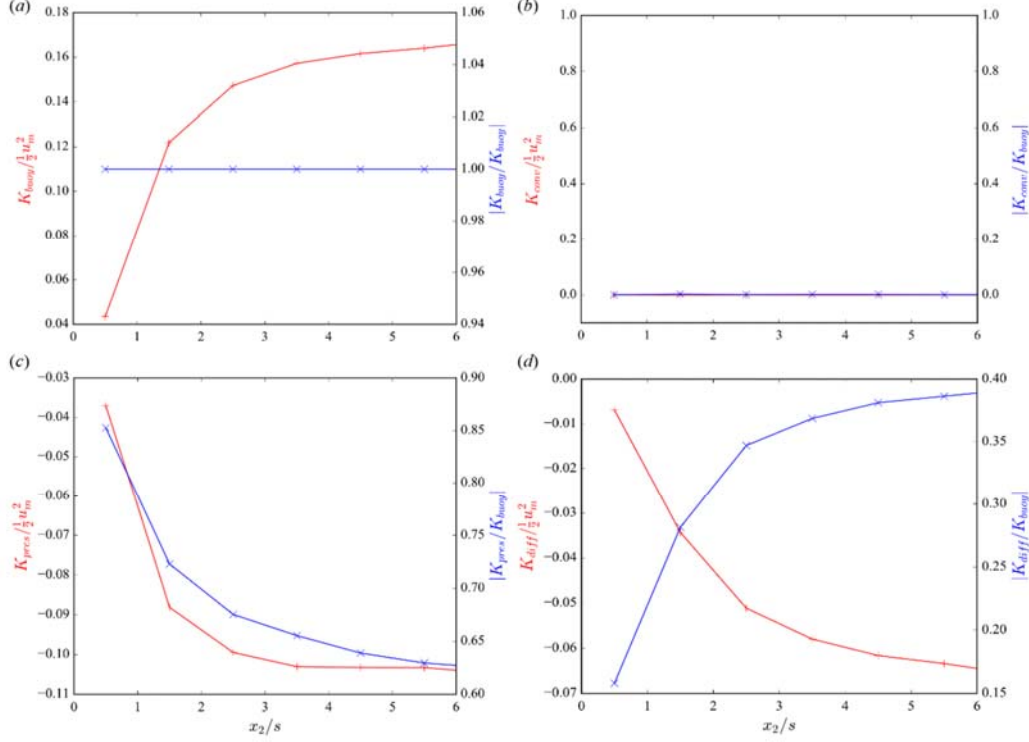


Figure 4. Distribution of the budget of the macroscopic kinetic energy $\langle \bar{K} \rangle^{x1}$ in the wall-normal direction. $s/d = 1.5$ ($\phi = 0.56$), $H/s = 20$ ($Da = 8.8 \times 10^{-6}$), $Ra = 20,000$, and $Sc = 250$.

Figure 5 shows the loss of the macroscopic kinetic energy due to the Darcy drag K_{Darcy} , (assuming that the superficial velocity calculated from the macroscopic simulation is identical to the DNS solution). K_{Darcy} is normalized by K_{mean} or K_{buoy} . It can be seen that K_{Darcy} is close to K_{buoy} in the region away from the wall ($x_2/s \gg 0$). However, K_{Darcy}/K_{buoy} is smaller than 0.85 in the first three REVs adjacent to the wall ($x_2/s < 3$). The DNS results confirm that the Darcy term, which accounts for the losses due to the Darcy drag, cannot account for all the losses of the macroscopic kinetic energy.

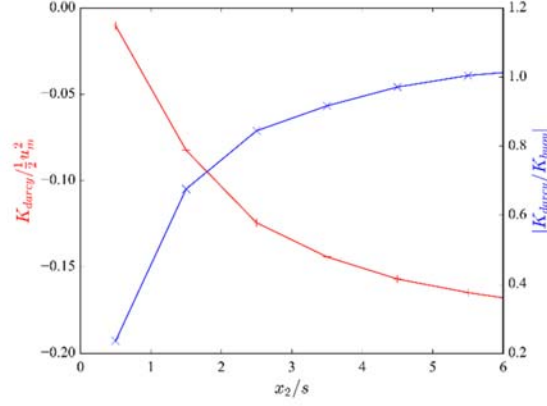


Figure 5. Distribution of the loss of the macroscopic kinetic energy $\langle \bar{K} \rangle^{x1}$ due to the Darcy drag. $s/d = 1.5$ ($\phi = 0.56$), $H/s = 20$ ($Da = 8.8 \times 10^{-6}$), $Ra = 20,000$, and $Sc = 250$.

The question arises of whether the difference between K_{Darcy} and K_{buoy} shown in figure 5 is because the Darcy number in the DNS case is not small enough. To answer this question, K_{pres}/K_{buoy} , K_{diff}/K_{buoy} , K_{conv}/K_{buoy} , and K_{darcy}/K_{buoy} in the first REV cell next to the bottom wall for different Darcy numbers are compared in figure 6. It is evident from this figure that all of these quantities stay almost constant as the Darcy number is decreased from 3.5×10^{-5} to 3.5×10^{-7} , suggesting that the Darcy numbers in our DNS cases are small enough for the presented analysis. The Darcy number has a noticeable effect as it is increased to $\sim 3 \times 10^{-5}$. In this case, K_{pres}/K_{buoy} and K_{Darcy}/K_{buoy} become smaller, K_{diff}/K_{buoy} becomes larger, whereas K_{conv}/K_{buoy} is still negligibly small. We speculate that higher K_{diff} at very large Darcy numbers is due to the mass dispersion, which is neglected in our macroscopic model (convection with very large Darcy numbers is out of the scope of this study).

Our budget analysis shows that, in the near wall region, there is a difference between the loss due to the Darcy drag K_{Darcy} and the overall loss which is identical to $-K_{buoy}$. Since the transport of $\langle \bar{K} \rangle^{x1}$ is negligibly small, this suggests that another source for the loss of $\langle \bar{K} \rangle^{x1}$ should be considered in the macroscopic equations.

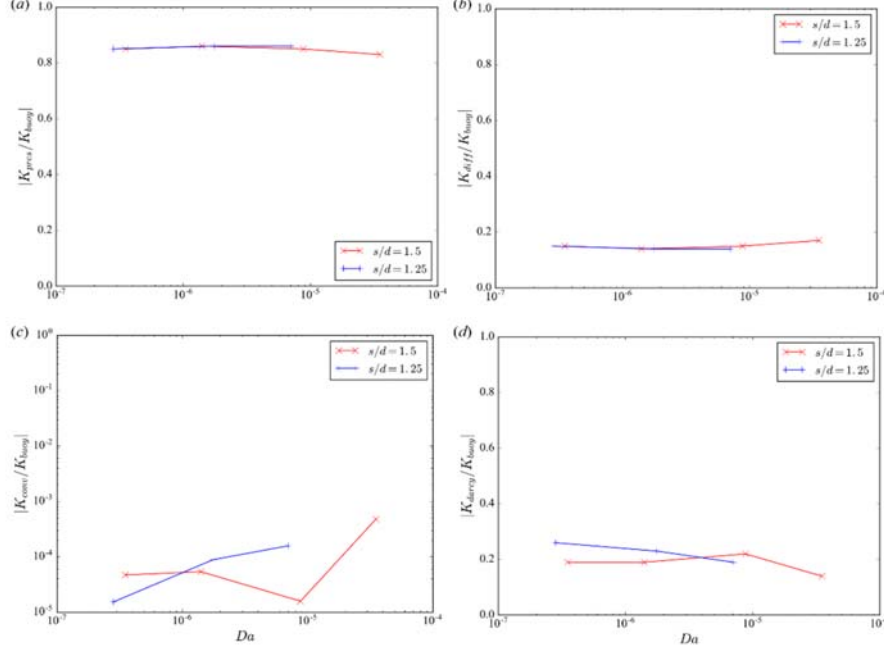


Figure 6. K_{pres}/K_{buoy} (a), K_{diff}/K_{buoy} (b), K_{conv}/K_{buoy} (c), and K_{Darcy}/K_{buoy} (d) in the first REV cell next to the bottom wall versus the Darcy number. $s/d = 1.5$ ($\phi = 0.56$) with $H/s = 10; 20; 50; 100$; and $s/d = 1.25$ ($\phi = 0.36$) with $H/s = 10; 20; 50$; $Ra = 20,000$; and $Sc = 250$.

4.2. Sh - Da dependence

According to our hypothesis, the macroscopic diffusion, the Darcy drag, and the buoyancy force are of the same order with respect to the Darcy number, so the macroscopic diffusion cannot be neglected even if the Darcy number is small. To examine our hypothesis, we investigated the relationship between the Sherwood number and the Darcy number. We varied Da in the range $3.5 \times 10^{-7} - 3.5 \times 10^{-5}$ for $s/d = 1.5$ ($\phi = 0.56$) and $2.8 \times 10^{-7} - 7 \times 10^{-6}$ for $s/d = 1.25$ ($\phi = 0.36$); the corresponding H/s ratios are in the range 10-100. If our hypothesis were true, the Sherwood number should gradually become independent of Da and should not approach the DOB solution.

The DNS results shown in figure 7 generally support our assumption, *i.e.* the values of Sh for $Sc = 250$ depend only weakly on Da , when Da is small enough. The values of Sh are also different from the DOB solution. As shown in figure 7(a), the same trend is found for $s/d = 1.25$ ($\phi = 0.36$) and $Sc = 1$, where Sh depends on Da for the cases with $s/d = 1.5$ ($\phi = 0.56$). The only exception is the case for $s/d = 1.5$ ($\phi = 0.56$) and $Sc = 1$, where Sh still increases with decreasing Da (but it is still far away from the DOB result). Test cases with even smaller Darcy numbers could be computed to probe the Da -dependence more thoroughly.

However, the calculation of these cases would be extremely expensive and hence out of the scope of this study.

It should be noted that the Darcy numbers for real applications are much smaller than the values used in the DNS cases. However, since our DNS results for the Sherwood number are approximately Da -independent, we expect that it is possible to predict the Sherwood numbers using DNS with relatively higher (computationally affordable) Darcy numbers.

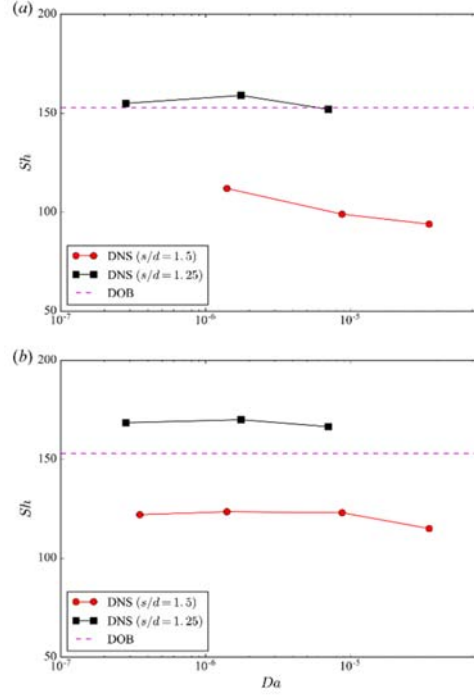


Figure 7. The $Sh(Da)$ dependence for the DNS and DOB cases. $s/d = 1.5$ ($\phi = 0.56$) with $\frac{H}{s} = 10; 20; 50; 100$; and $s/d = 1.25$ ($\phi = 0.36$) with $H/s = 10; 20; 50$; and $Ra = 20,000$. (a) $Sc = 1$ and (b) $Sc = 250$.

In a recent DNS study, Liu *et al.* (2020a) proposed the following correlation for estimating the Nusselt number (equivalent to Sh in this study):

$$Sh \approx c \cdot \phi \left(\frac{H}{l} \right)^4 Sc^2 Re_{rms}^2 Ra_f^{-1} + 1, \quad (23)$$

where c is an undetermined constant according to the work of Grossmann & Lohse (2000; 2001; 2004), l is the minimum spacing between the obstacles, $Re_{rms} = U_{rms}l/\nu$ is the Reynolds number based on the volume-averaged r.m.s. velocity magnitude, and $Ra_f = H^3\beta\Delta cg/\nu D_f$ is the Rayleigh number defined for the free fluid flow. If we set the value of c to 1250 and determine U_{rms} from our DNS results, the results of equation (23) are in good agreement with

our DNS results for different values of ϕ and Sc , see figure 8. **Error! Reference source not found.** It should be noted that equation (23) is proposed based on the flow condition that viscosity dominates, hence intense kinetic energy dissipation takes place within the bulk domain and turbulence is suppressed in the pore canals. For the volume and time averaged kinetic energy dissipation rate $\langle \epsilon_u \rangle^{v,t}$ the following proportionality is valid: $\langle \epsilon_u \rangle^{v,t} \sim \phi \nu U_{rms}^2 / l^2$, which corresponds to the ∞ -regime of classical Rayleigh Bernard convection (without porous media) introduced by Grossmann & Lohse (2001) for large Sc and small Ra_f . A good agreement between predictions obtained using equation (23) and our DNS results indicates the significance of macroscopic diffusion in momentum transport.

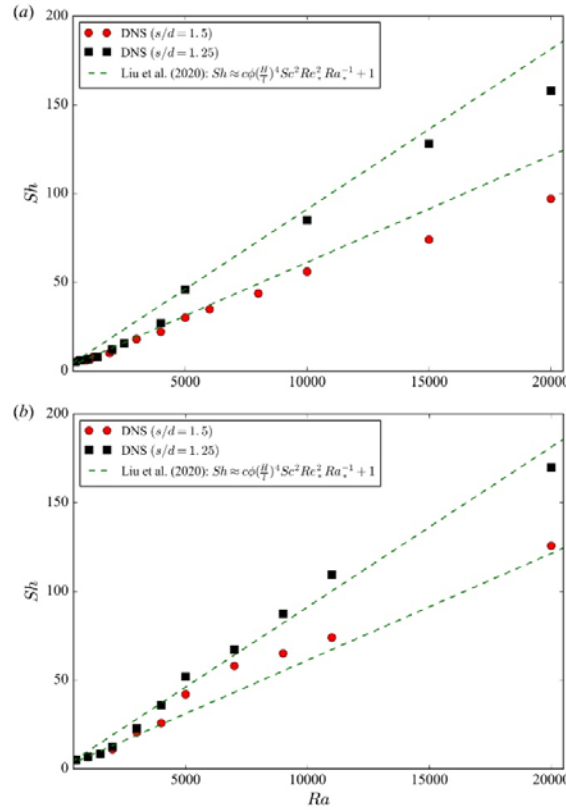


Figure 8. Sherwood number versus the Rayleigh number for Ra in the range 500 – 20,000 compared to the correlation proposed by Liu *et al.* (2020a). (a) $Sc = 1$, (b) $Sc = 250$.

5. Macroscopic modeling results

Since the leading order terms of Da for diffusion are accounted for in the TLSD model, this model can be used in principle to calculate cases characterized by small Darcy numbers. In this section, we test whether and how the model results approach the DNS results as $Da \rightarrow 0$. In addition, we investigate the range of parameters for the validity of the TLSD model.

5.1. Sherwood number

Figure 9 shows the relationship between the Sherwood number and the Rayleigh number when H/s is 20 and Rayleigh numbers are up to 20,000. The results of our macroscopic model are compared with the correlation obtained from the DNS results (see Gasow *et al.* 2020) as well as the DOB results. In the DNS, Sh depends on s/d or ϕ for $Ra > 2,000$. The value of Sh increases as s/d or ϕ decreases, while the difference becomes larger as Ra increases. In the large Rayleigh-number regime ($Ra \geq 5,000$), the $Sh = f(Ra)$ scaling changes from a linear scaling $Sh \sim Ra$ for $s/d = 1.25$ ($\phi = 0.36$) to a non-linear scaling $Sh \sim Ra^{0.8}$ for $s/d = 1.5$ ($\phi = 0.56$). These characteristics are not captured in DOB simulations but are well reproduced in our macroscopic model simulations.

For the current small H/s (or Da) value, both DNS and model results show that the Sherwood number increases as the Schmidt number is increased from 1 to 250. Similar to $Sc = 1$, the scaling for $Sc = 250$ changes from linear ($Sh \sim Ra$) for $s/d = 1.25$ ($\phi = 0.36$) to non-linear ($Sh \sim Ra^{0.8}$) for $s/d = 1.5$ ($\phi = 0.56$). This behavior is reproduced in our macroscopic model solution, whereas the effect of the Schmidt number is not accounted for in the DOB equations.

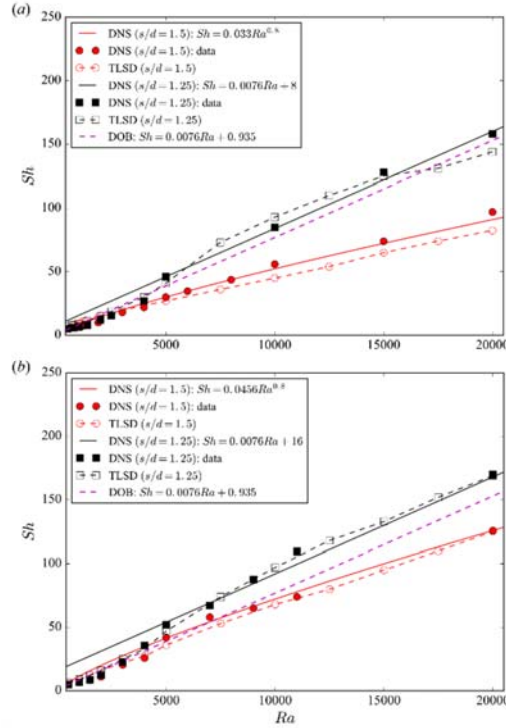


Figure 9. Sherwood number versus the Rayleigh number with Ra in the range 500 – 20,000 and $H/s = 20$ for three values of the Darcy number: $Da = 8.8 \times 10^{-6}$ ($s/d = 1.5$), $Da = 5.4 \times 10^{-6}$ ($s/d = 1.4$), and $Da = 1.8 \times 10^{-6}$ ($s/d = 1.25$). (a) $Sc = 1$, (b) $Sc = 250$.

We neglected the high order terms with respect to Da when we proposed the TLSD model. However, since the leading order term with respect to Da is kept in the momentum equation (11), the effect of Da on natural convection can still be accounted for when its value is small enough. The relationship between the Sherwood number and the Darcy number for $Ra = 20,000$, $Sc = 1$ or 250 , and $s/d = 1.5$ or 1.25 ($\phi = 0.56$ or 0.36) is shown in figure 10. The macroscopic model results are compared with the DNS results as well as with the DOB results. Recall that a_v^* is only related to the pore scale geometry and is independent of the flow conditions and the Darcy number. The macroscopic model simulations are in good agreement with the DNS for $Da \leq 2 \times 10^{-6}$ and for different Schmidt numbers. By contrast, the DOB results are independent of the Darcy and Schmidt numbers. The DOB simulations overpredict the Sherwood number for $s/d = 1.5$ ($\phi = 0.56$) and underpredict Sh for $s/d = 1.25$ ($\phi = 0.36$).

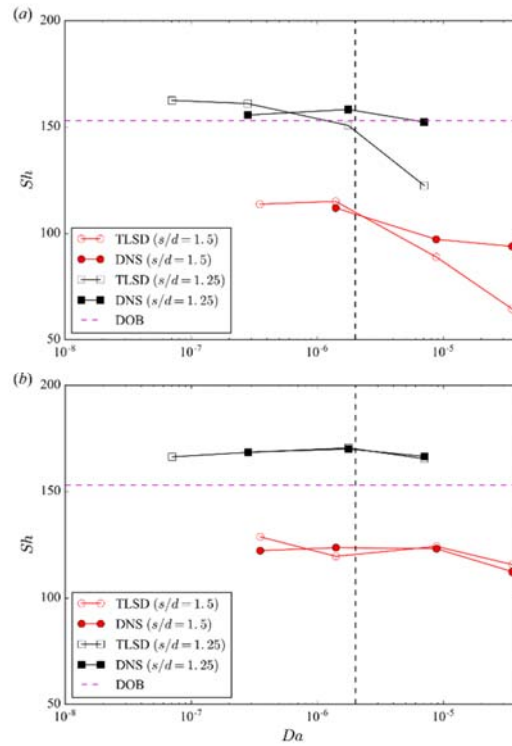


Figure 10. Sherwood number versus the Darcy number. $s/d = 1.5$ ($\phi = 0.56$) with $H/s = 10; 20; 50; 100$; and $s/d = 1.25$ ($\phi = 0.36$) with $H/s = 10; 20; 50$; and $Ra = 20,000$. (a) $Sc = 1$ and (b) $Sc = 250$.

5.2. Species concentration and velocity statistics

The vertical profiles of the temporally and horizontally averaged macroscopic quantities (time- and line-averaged species concentration $\langle \bar{c} \rangle^{x1}$, species concentration fluctuation $\langle \hat{c}^{rms} \rangle^{x1}$, and velocity fluctuations $\langle \hat{u}_1^{rms} \rangle^{x1}$ and $\langle \hat{u}_2^{rms} \rangle^{x1}$) for $s/d = 1.5$ ($\phi = 0.56$) and $Sc = 1$ are shown in figure 11. The Rayleigh numbers are 5,000 and 20,000. The results of our macroscopic TLSD model are compared with the DNS results as well as the DOB results. It is evident that our macroscopic TLSD modeling results for $\langle \bar{c} \rangle^{x1}$, $\langle \hat{u}_1^{rms} \rangle^{x1}$, and $\langle \hat{u}_2^{rms} \rangle^{x1}$ are more accurate than the DOB results in the first REV next to the wall. The DNS results show that all statistical results can be well scaled by the pore size s and that the influence of the bounding walls is limited to within the first three REV's next to the wall (Gasow *et al.* 2020). Thus, the boundary layer thickness is determined by the pore size s , instead of the Rayleigh number, as suggested in Huppert & Neufeld (2014). These features are all well captured in the macroscopic simulation.

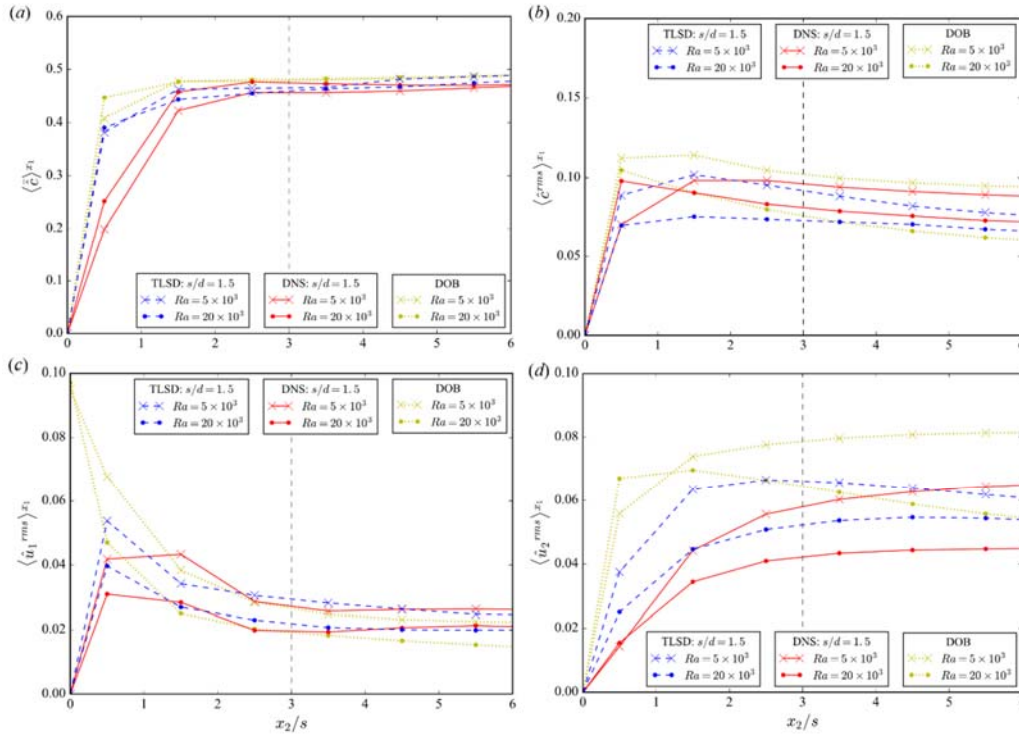


Figure 11. The vertical profiles of the temporally and horizontally averaged macroscopic quantities for $s/d = 1.5$ ($\phi = 0.56$), $H/s = 20$ ($Da = 8.8 \times 10^{-6}$), and $Sc = 1$. The Rayleigh number Ra is varied. The distance from the wall is normalized by the pore size s . (a) Time- and line-averaged species concentration $\langle \bar{c} \rangle^{x1}$, (b) root mean square (r.m.s.) of the species concentration fluctuation $\langle \hat{c}^{rms} \rangle^{x1}$, (c) streamwise velocity fluctuation $\langle \hat{u}_1^{rms} \rangle^{x1}$, (d) wall-normal velocity fluctuation $\langle \hat{u}_2^{rms} \rangle^{x1}$.

The same statistical quantities are shown in figure 12 for $Sc = 250$. It can be seen that these statistical quantities are only marginally changed when a much higher Schmidt number is used in the simulation. Similar to the results for $Sc = 1$, the macroscopic modeling results are also in good agreement with the DNS results. The macroscopic TLSD model simulation predicts higher mass concentration $\langle \bar{c} \rangle^{x1}$ in the first REV next to the wall and higher transverse velocity fluctuation $\langle \hat{u}_2^{rms} \rangle^{x1}$. One possible reason for this discrepancy is that the neglected high order terms with respect to Da may lead to modeling errors in the boundary layer region. The TLSD model accuracy can be further improved by decomposing the flow domain into a boundary layer region and a central region, so the modeling in the boundary layer region can be improved. However, this would make the model more complicated and difficult to apply. This modeling approach is not adopted in our study to achieve a compromise between the accuracy and simplicity of the macroscopic model.

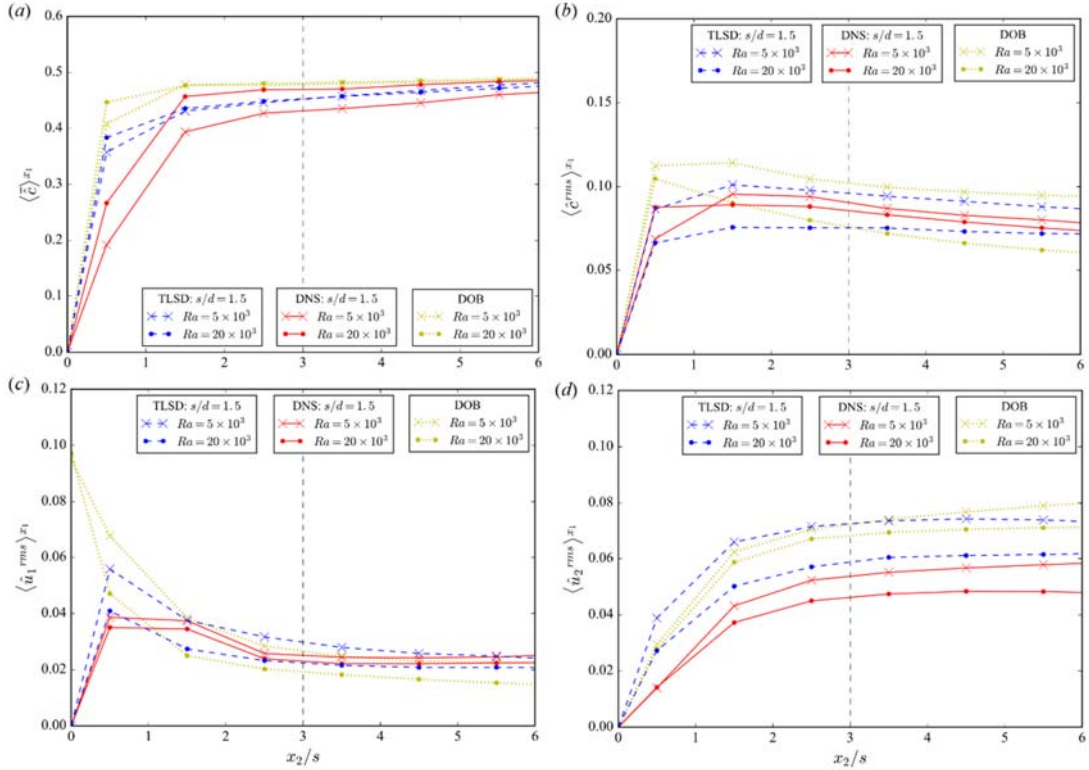


Figure 12. The vertical profiles of the temporally and horizontally averaged macroscopic quantities for $s/d = 1.5$ ($\phi = 0.56$), $H/s = 20$ ($Da = 8.8 \times 10^{-6}$), and $Sc = 250$. The Rayleigh number Ra is varied. The distance from the wall is normalized by the pore size s . (a) Time- and line-averaged species concentration $\langle \bar{c} \rangle^{x1}$, (b) r.m.s. of the species concentration fluctuation $\langle \hat{c}^{rms} \rangle^{x1}$, (c) streamwise velocity fluctuation $\langle \hat{u}_1^{rms} \rangle^{x1}$, and (d) wall-normal velocity fluctuation $\langle \hat{u}_2^{rms} \rangle^{x1}$.

5.3. Transient macroscopic fields

To validate the results of our macroscopic TLSD model, we first compare the transient flow fields obtained from macroscopic simulations of equations (10)-(12) with those obtained from the DNS results discussed in the previous section and the DOB simulations reported in Gasow *et al.* (2020). For this purpose, the velocity field and the species concentration obtained with the macroscopic simulations and the DNS were volume-averaged (over each REV).

The distribution of the instantaneous $Re_K = (|\mathbf{u}|K^{1/2})/\nu$ for $Ra = 20,000$, $s/d = 1.5$ ($\phi = 0.56$), $H/s = 100$, and $Sc = 250$ is shown in figure 13. The macroscopic TLSD solution (figure 13(c)) is qualitatively similar to the DNS solution (figure 13(a)) and DOB solution (figure 13(b)). Both the DNS solution and macroscopic solutions indicate that the local Reynolds number is $Re_K < 4 \times 10^{-3}$. This shows that the studied parameter range is well in the Darcy regime ($Re_K \ll 1$), hence the Forchheimer term in the momentum equation can be safely neglected. Despite the laminar flow in the pore scale, the macroscopic flow field is transient and chaotic. However, the strong spatial variation of the velocity field obtained from the DNS is neither captured in TLSD nor in the DOB simulations.

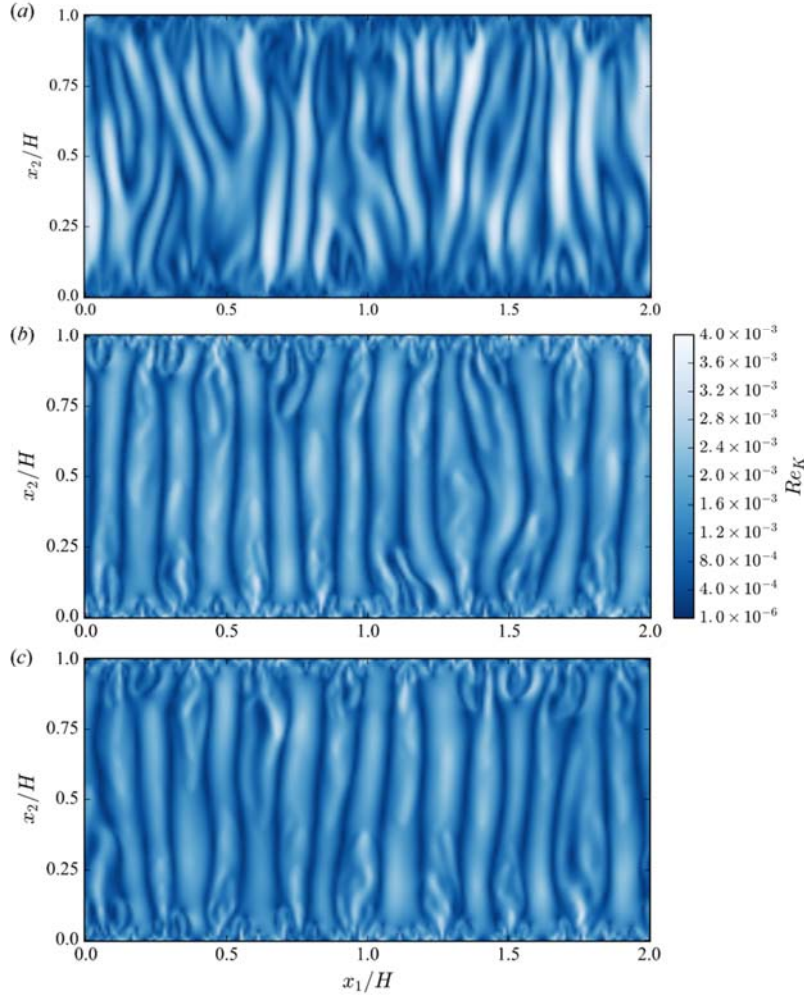


Figure 13. Instantaneous volume averaged Reynolds number Re_K , $H/s = 100$ ($Da = 3.5 \times 10^{-7}$), $s/d = 1.5$ ($\phi = 0.56$), $Ra = 20,000$, and $Sc = 250$. (a) DNS, (b) DOB, (c) TLSD.

Snapshots of the instantaneous species concentrations for $H/s = 100$, $s/d = 1.5$ ($\phi = 0.56$), $Ra = 20,000$, and $Sc = 250$ are shown in figure 14. The DNS solution (figure 14(a)), TLSD solution (figure 14(c)), and the DOB solution (figure 14(b)) all exhibit large mega-plumes structures in the internal region and small proto-plumes in the boundary layers. They occur due to the rising of a fluid with low species concentration and the sinking of a fluid with high species concentration, forming the instabilities in the boundary layer region (Hewitt *et al.* 2012; Kränzien & Jin 2019).

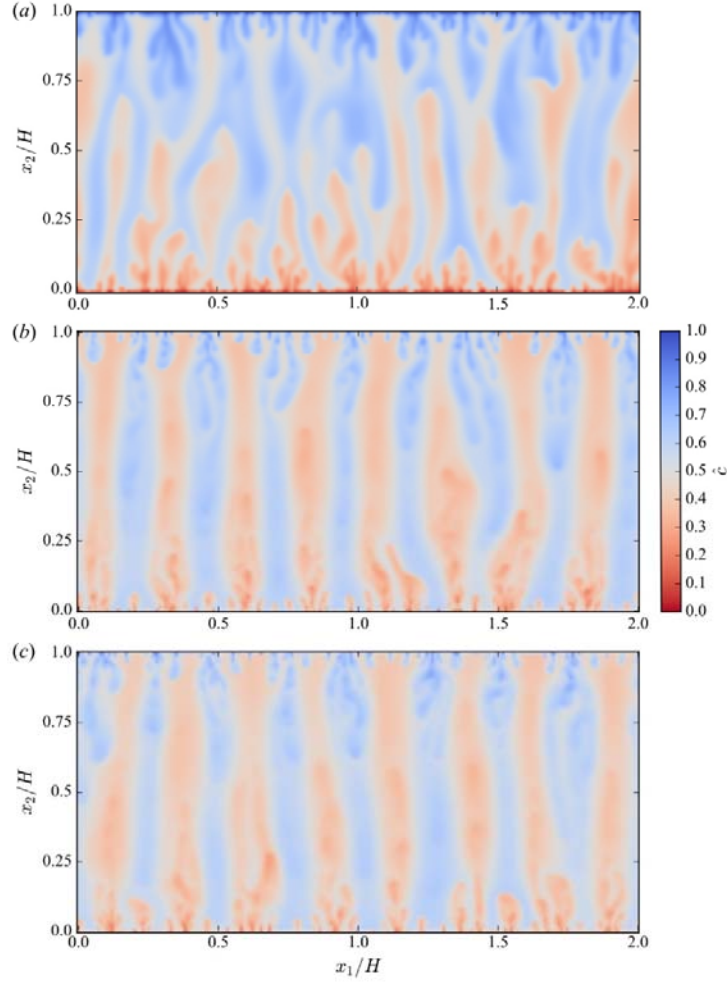


Figure 14. Instantaneous volume-averaged species concentration $\hat{c}$, $H/s = 100$ ($Da = 3.5 \times 10^{-7}$), $s/d = 1.5$ ($\phi = 0.56$), $Ra = 20,000$, and $Sc = 250$. (a) DNS, (b) DOB, (c) TLSD.

While the macroscopic TSDL model and DOB solution exhibit relatively regular mega plumes, in the DNS solution the mega-plumes are more irregular and chaotic. A possible reason is that the Darcy number in our simulation is still not small enough, while the TLSD model is proposed for problems with low Darcy numbers. The transient flow field from macroscopic simulation converges slower than the Sherwood number and other statistical results with decreasing Da .

The DNS study reported in Gasow *et al.* (2020) shows that the number of mega-plumes increases with the decrease of Da . Figure 15 shows the time-averaged Fourier transform of the dimensionless mass concentration $\hat{c}$ along the centerline at $x_2 = H/2$. The peak wave number calculated from the TLSD simulation is still higher than the DNS result, but it is lower than the DOB result. Figure 16 shows that the peak wave number from DNS approaches the TLSD or DOB results as the Darcy number approaches 0. However, DNS of natural convection with

smaller Darcy numbers are still needed to confirm that the peak wave number from DNS will not exceed the TLSD results and approach the DOB results.

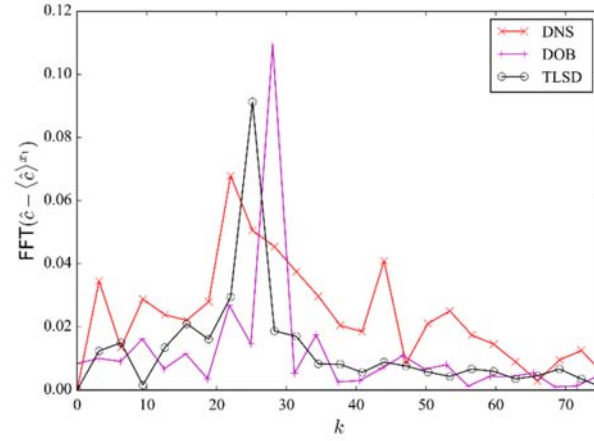


Figure 15. Average spectra of the dimensionless mass concentration, $\hat{c}$ of the DNS, DOB, and TLSD results at mid-height $x_2 = H/2$, $H/s = 100$ ($Da = 3.5 \times 10^{-7}$), $s/d = 1.5$ ($\phi = 0.56$), $Ra = 20,000$, and $Sc = 250$.

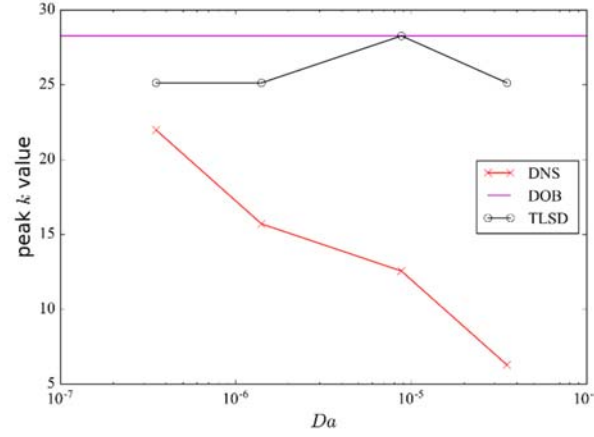


Figure 16. Peak wavenumber k for the mega-plumes of the DNS, DOB, and TLSD results for different Darcy numbers with $s/d = 1.5$ ($\phi = 0.56$), $Ra = 20,000$, and $Sc = 250$.

The 3-dimensional DOB simulations by Pirozzoli *et al.* (2021) revealed the supercells at the boundary, which are the footprints of megaplumes dominating the interior part of the flow. They suggest that these supercells might lead to the nonlinear scaling of $Sh(Ra)$ in the ultimate regime of high Ra numbers. Future work needs to investigate whether these supercells will be also captured by 3-dimensional TLSD simulations. Elucidating how macroscopic diffusion affects the plume structures is also a subject of future investigation.

6. Conclusions

The DNS results of Gasow *et al.* (2020) (extended in this study) show that the pore-scale geometry also has significant effects on natural convection in porous media, in particular, the boundary layer thickness is determined by the pore size instead of the Rayleigh number. Based on this, we have proposed the following TLSD model: we assume that pore scale structures affect the momentum transport through macroscopic diffusion. The macroscopic diffusion is of the same order with respect to the Darcy number as the Darcy drag and the buoyancy force, thus, it cannot be neglected even if the Darcy number is small. It is determined by two length scales, the pore size characterized by $\sqrt{K}$ and the distance between the lower and upper boundaries H .

The DNS results show that the loss of the macroscopic kinetic energy is mainly due to microscopic diffusion and the pressure gradient. The loss captured in Darcy's law is only part of the overall loss, even if the superficial velocity is accurately calculated in the DOB equation. The macroscopic diffusion term added here to the momentum equation accounts for the additional loss of the macroscopic kinetic energy. Our DNS results also show that the Sherwood number is almost independent of the Darcy number when the Darcy number is small enough. Thus, the diffusion term is of the same order of the Darcy number ($Da = K/H^2$) as the buoyancy force term and the Darcy term.

A new macroscopic model for simulating natural convection in porous media is developed based on the TLSD assumption. The results of our model are validated extensively by comparison with the DNS as well as the DOB results. The comparison shows that the new macroscopic model performs well as long as $Da \leq 2 \times 10^{-6}$. Simulations of the model predict a much more accurate Sherwood number, r.m.s. mass concentration, and r.m.s. velocity than simulations that employ the DOB equations. They also predict the structures of mega-plumes and proto-plumes with reasonable accuracy. In particular, the new model results show that the $Sh = f(Ra)$ scaling changes from a linear scaling to a non-linear scaling as the porosity increases. If the Rayleigh number and Darcy number are fixed, the Sherwood number increases with the increase of the Schmidt number and the decrease of the porosity. These trends agree with the DNS results, whereas they cannot be captured by the DOB simulations. We expect that these trends, as well as the TLSD assumption, also apply to 3-dimensional flows. However, how macroscopic diffusion affects the plume structures remains an open question.

Some discrepancies between the new macroscopic modeling results and the DNS results can be found in the boundary layer. The new macroscopic model over-predicts the mean mass

concentration in the first REV next to the wall. This error may be reduced if the higher order terms with respect to Da , e.g., the mass dispersion, are considered. However, the current macroscopic model appears preferable due to its simplicity.

This work is the first step towards modeling fundamental issues arising at the pore scale in CO₂ sequestration processes. However, it should be noted that a real CO₂ sequestration process is much more complicated. It has been extensively investigated by numerical modeling in the last two decades (Weir *et al.* 1995, 1996; Lindeberg & Wessel-Berg 1997; Hassanzadeh *et al.* 2005, 2007; Bickle *et al.* 2007; Pruess & Zhang 2008; Chen *et al.* 2018). It is characterized by three-dimensional, inherently transient multiphase flow with much more complicated pore-scale geometries and much lower Darcy numbers than those studied in this research (Michael *et al.* 2010; Riley 2010; Huppert & Neufeld 2014).

Appendix A1 Momentum dispersion and species concentration dispersion

Breugem *et al.* (2006) argued that momentum dispersion has negligible effects on convection in porous media. This agrees with the study by Rao *et al.* (2020), who showed numerically that momentum dispersion should be accounted for only when the local Reynolds number $Re_K \gg 1$. Hence, the momentum dispersion is neglected here as well.

The effects of mass dispersion (or thermal dispersion for heat transfer problems) have been extensively studied in recent years, as discussed in the Introduction (Fahs *et al.* 2020; Alomar 2019; Wen *et al.* 2018; Liang *et al.* 2018). The dispersion term in equation (7) is often modeled using a Fickian dispersion tensor, first introduced by Bear (1961) and expressed as:

$$\phi \langle u_i c \rangle^i = D_{ij} \frac{\partial \check{c}}{\partial \check{x}_j}, \quad (24)$$

where the dispersion tensor D_{ij} is calculated as:

$$D_{ij} = (\alpha_l - \alpha_t) \check{u}_i \check{u}_j / |\check{\mathbf{u}}| + \alpha_t \delta_{ij} |\check{\mathbf{u}}|, \quad (25)$$

and α_l and α_t are the longitudinal and transverse diffusivities, respectively. They can be determined from the numerical results for the flow and mass transfer in a REV with a linear concentration gradient in the streamwise or transverse direction, see Nakayama & Kuwahara (1999) and Pedras & de Lemos (2008). These studies suggest that D_{ij} has the scaling of the form $D_{ij} \sim Pe_K^\eta$, where the local Peclet number Pe_K is defined as

$$Pe_K = Re_K Sc = \frac{|\bar{\mathbf{u}}|}{u_m} Re_m Da^{1/2}, \quad (26)$$

where $Re_K = |\bar{\mathbf{u}}|\sqrt{K}/\nu$ and $Re_m = u_m H/\nu$ are the local and global Reynolds numbers, respectively. Delgado (2007) and Nakayama & Kuwahara (1999) suggested that the scaling coefficient n is between 1 and 2. As a consequence, D_{ij} is expected to be of order between $Da^{1/2}$ and Da , while D_m is of order Da^0 . When the Darcy number is small enough, $|D_{ij}| \ll D_m$. Since we are interested in natural convection with small Darcy numbers, we only retain the leading order terms with respect to Da for diffusion in equation (7). Thus, due to this theoretical derivation, mass dispersion can also be neglected.

The dispersion at the macro-scale (macrodispersivity) suggested by Gelhar *et al.* (1992), Lallemand-Barres & Peaudecerf (1978), Neuman (1990), and Liang *et al.* (2018) is not considered in this study since its effect on the plume scale has not yet been fully elucidated (Zech *et al.* 2019). Instead, the effect of dispersion is modeled as macroscopic diffusion in the momentum equation. The macroscopic diffusion affects the velocity field and then accounts for the dispersion in the species concentration indirectly.

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Declaration of Interests.

The authors report no conflict of interest.

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