Continuous upscaling of the 3D diffusion equation in a heterogeneous medium

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HIGHLIGHTS

- A theory for continuous multiscale upscaling of 3D flows is developed.
- General equations for upscaling of densities and fluxes are obtained.
- Upscaling of the steady-state diffusion process in a heterogeneous medium is studied.
- An equation for upscaling the diffusion coefficient is obtained for a large class of solutions.
- Depending on a solution, an upscaling equation for the diffusion coefficient may be different.

ARTICLE INFO

ABSTRACT

We describe the procedure of continuous upscaling of the flow equations. Multiscale averaging is a necessary operation in many applications. Under the introduction of the continuous line of scales, upscaling becomes similar to a Markov process described by a partial differential equation of the Ornstein-Uhlenbeck type. While previously [Shapiro, Chem. Eng. Sci. 234, 116454] the procedure of continuous upscaling was formulated for 1D flows, here we generalize it onto 3D processes. For upscaling 3D fluxes, new laws are formulated. The procedure is applied to upscale the steady-state diffusion (or heat conduction, or the pressure equation) in a heterogeneous medium. Rules for upscaling the diffusion coefficient are derived. In many cases, the upscaling of the diffusion coefficient may depend on the chosen class of solutions. It is studied numerically. The lowest values of the coefficient and the zones of its sharp variation contribute most to upscaling.

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1. Introduction

1.1. Statement of the problem

Upscaling is a common operation in many areas of fundamental and applied science involving multiple scales. It is applied in the theory of turbulence (Monin and Yaglom, 1971; Germano, 1992; Catton, 2006), microfluids and colloids (Eringen, 1964; Shapiro, 1996, Krehel et al., 2015), multiphase flows (Cushman, 1985; Faghri and Zhang, 2020), flows in porous media (Gray and Miller, 2014). Typical industrial applications are for ecology and groundwater flows (Baveye and Sposito, 1985), petroleum reservoir engineering (Bedrikovetsky and Rowan, 1993), chemical and biochemical engineering (Quintard and Whitaker, 1993; De Los Santos-Sánchez et al., 2016; Yang et al., 2015; Travkin and Catton, 2001). Depending on a tradition in a particular discipline, the upscaling operation may also be called "averaging", "homogenization", "spatial filtering", or "coarse-graining" (Municchi and Icardi, 2020).

The goal of upscaling is to derive the flow equations and expressions for transport coefficients on a coarser scale, starting from the description of the flow and geometry of the medium on a finer scale. More generally, the goal is to establish the connection between, seemingly, different physical descriptions of the processes on the different scales and, by that, to validate the application of a simpler and more practically useful macroscale description.

An overview of the previously developed methods for upscaling is contained in our previous paper (Shapiro, 2021). These methods may be divided into general and problem-specific. The problem-specific methods utilize peculiarities of a particular system on the microscale, while the general methods may virtually be applied to any such system. Obviously, this classification does not tell any-

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List of designations

$\boldsymbol{a}$	Averaging operator (without scaling)
$b(s)$	Displacement coefficient
c	Concentration
$d(\boldsymbol{x})$	Diffusion coefficient
D	Dimension of the space
$D(s)$	Averaging (dispersion) coefficient
E_s	Substitution variable
G_s	Rescaled diffusion coefficient
H	Constants in the factorized solution
$\boldsymbol{I}$	Unity operator
$\boldsymbol{j}$	Flux
K	Number of the discretization points in the numerical solution
l	Transfer density
$\boldsymbol{L}$	Non-convective flux
m, n	Densities of arbitrary physical measures (mass, momentum, energy...)
q	Source term in a transport or diffusion equation
s	The scale variable (minus logarithm of the scaling factor)
$\boldsymbol{S}$	Scaling operator (without averaging)
t	Time
$\boldsymbol{u}$	Infinitesimal upscaling operator

$\boldsymbol{U}$	Complete upscaling operator (both scaling and averaging)
$\boldsymbol{v}$	Velocity
x	Coordinate for a 1D solution
$\boldsymbol{x} = (x_1, x_2, x_3)$	Vector of coordinates
y	Substitution variable
Z	Transformed component of the diffusive flux
α	Scaling factor
$\beta(s)$	Constant of integration in the upscaling equation
Γ	Measurable set
ε	Parameter in the initial condition
μ	Measure
ρ	Mass density
τ	Operator of exact scaling by time
ξ	Operator of exact scaling by a spatial coordinate

Subscripts and superscripts

$i, j, k \cdots$	Lowercase indices, by which the summation is always assumed
$I, J, K \cdots$	Uppercase indices, by which the summation is never carried out
s	Upscaled value (dependent on s)

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thing about the theoretical or practical significance of an upscaling method; for example, such a fundamental discipline as statistical mechanics may, from a certain point of view, be regarded as a problem-specific method of upscaling, since its goal is to derive macroscopic properties of a matter exceeding from known interactions on the molecular scale. There were multiple attempts to extend the methods of statistical mechanics and gas kinetic theory from molecular to disperse colloidal systems, like particles in porous media (e.g. Shapiro, 1996; Bedrikovetsky, 2008). However, these works still impose rather strong restrictions onto the behavior of the systems on micro-level.

Among the general methods, the volume-averaging theory (VAT), in different versions, is the most widely applied (Whitaker, 1999; Travkin and Catton, 2001; Gray and Miller, 2014). A more general upscaling procedure is weighted averaging (Marle, 1967, 1982; Cushman, 1984a,b, 1985; Kalaydjian, 1987; Quintard and Whitaker, 1994a,b,c). The theory of homogenization may also be regarded as general, although it is commonly applied to spatially periodic media (Arbogast et al., 1990; Allaire and Raphael, 2007; Panfilov and Rasoulzadeh, 2013; Rasoulzadeh et al., 2014; Bloch and Auriault, 2019; Municchi and Icardi, 2020). It should be remarked that an assumption about spatial periodicity on the microscale is often assumed also in the VAT, to derive the closure relations for the macroscale equations (Ryan et al., 1980; Allaire and Raphael, 2007).

A specific feature of the general upscaling methods is that the transition between the scales is asymptotic, in the limit of a large ratio of the characteristic scale units. A substantial effort is required to make this transition mathematically strict and physically sound (see, e.g., Gray, 1982; Cushman, 1984a,b; Nguetseng, 1989; Hassanizadeh and Gray, 1990; Allaire and Briane, 1996; Baveye, 2004). This problem becomes especially difficult if the transitions between multiple scales are involved, as usually required for the flows in porous media (Cushman, 1985; Allaire and Briane, 1996; Gray and Miller, 2014). The natural porous media, involved e.g. in carbon dioxide storage, geothermal energy

engineering, or petroleum production, are heterogeneous on multiple scales, with a continuous range of possible heterogeneities from micro- to geological scale. Description of such porous media by "jumpwise" transitions between the scales may require rather specific assumptions about the structures at each scale. Moreover, the interaction between the scales may result in a nonlocal character of the resulting macroscale equations. The effect of scale interaction may be accumulated under multiscale transitions, resulting in the appearance of the nonlocal transport equations containing the kernels with long "tails" (Panfilov and Rasoulzadeh, 2013; Rasoulzadeh et al., 2014).

While the mathematics of upscaling deals mainly with jumpwise, single-step transitions between the scales, the physical intuition makes it more appealing to consider a continuous transition, passing all the intermediate scales. The theory of continuous upscaling may be required in a number of engineering applications, like flows in natural porous media or the turbulent flows. Such a theory was developed in our previous paper (Shapiro, 2021). We introduced a continuous line of scales s, such that the difference $s_2 - s_1$ corresponds to the minus logarithm of the scaling coefficient between scales 1 and 2. It was postulated that the composition of upscalings between subsequent scales is an upscaling so that the operators of upscaling form a continuous semigroup parametrized by s. The line of scales served as a new dimension, and physical values evolved along this new coordinate. The evolution was described by a partial differential equation, similar to the Ornstein-Uhlenbeck equation from the theory of continuous-time Markov processes (Gardiner, 1985). This equation is well studied and solved, so that the theory of continuous upscaling may, in a way, be more straightforward than the VAT, the weighted averaging, or the homogenization theory. Another simplification is that the problem of multiscale transitions is non-existent anymore since such transitions form the basis of the whole upscaling scheme. It was shown that, under some obvious physical assumptions, the suggested method of continuous upscaling is unique, within the values of some coefficients.

The present paper is developing the previous work in the two directions. The theory of continuous upscaling was previously formulated for one-dimensional densities and fluxes. In the first part of the present work (Section 2), it is generalized onto three dimensions. This generalization is almost trivial for densities of physical values (like mass, momentum, or energy density); however, it requires some re-consideration for the fluxes. While in 1D the fluxes were shown to obey the same upscaling laws as densities, the situation is different in 3D, where upscaling of the fluxes obeys more complex relations.

The second part of the paper (Section 3) discusses upscaling of the 3D steady-state diffusion equation. This equation is of great practical importance, because of its wide range of applications. Additionally, a similar mathematical formulation describes, apart from diffusion, heat conduction in heterogeneous bodies and the flow of a compressible liquid in a porous medium.

The diffusion equation has been a good sample test problem for the different methods of upscaling. There is an overwhelming amount of literature discussing upscaling of this equation and its generalizations in the framework of VAT (Ryan et al., 1980; Whitaker, 1999; Valdés-Parada and Alvarez-Ramírez, 2011; Valdes-Parada et al., 2010, 2011, 2017), weighted averaging (Quintard and Whitaker, 1993), and the homogenization theory (Allaire and Raphael, 2007; Panfilov and Rasoulzadeh, 2013; Rasoulzadeh et al., 2014). In particular, the above literature discusses the interaction of the scales, the effect of surface chemical reactions on the effective diffusivity, and the non-local character of the macroscopic averaged equations.

In the present work, we prefer to keep the form of the upscaled diffusion equation and attempt to produce the upscaling equation for the diffusion coefficient. We show that this is possible in the 1D case. However, in 3D the upscaling equation for the diffusion coefficient does not seem to always exist, and, even if existing, may be different for the different classes of solutions of a diffusion equation. As a basic example, we consider the case where the diffusion coefficient may be factorized on the original scale. Even this case turns out to be nontrivial and producing the different upscaling equations for the diffusion coefficient. One of these equations, corresponding to a wide class of solutions, is investigated numerically. It is demonstrated that under transition from a finer to a coarser scale, the diffusion coefficient converges to a constant value, which is close to a minimum value of the diffusion coefficient on the original scale.

1.2. Agreements about designations

We consider upscaling in a region of a 3D space parametrized by Cartesian coordinates $\boldsymbol{x} = (x_1, x_2, x_3)$. The vectors in this space will be denoted by bold letters, while their components by the same letters with subscripts or superscripts. The bold letters will also be reserved for the operators.

A widely used agreement in mathematical notation is the Einstein tensor notation: the summation by an index is implied if this index appears twice in a monomial, once as a superscript, and once as a subscript. Since in this work the coordinate transformations and the chain rule are not extensively applied, it is more convenient to apply a different convention. We assume, simply, that *any lowercase index participates in summation, while any uppercase index is standalone*. For example, the expression $\frac{\partial T^{kL}}{\partial x_{iL}}$ presumes summation over indices i and k, while index L is fixed. Even in the expressions like $\frac{\partial^2 c}{\partial x_j^2}$, summation by j is assumed, while $\frac{\partial^2 c}{\partial x_J^2}$ represent a single term for a specific value of J. As e.g. an example of Appendix B shows, this agreement serves to a great simplification of, otherwise, even more cumbersome computations.

Any upscaling operation should be applied to a set of partial differential equations expressing the balances of some physical values. A general form of a balance equation is

$$\frac{\partial m_s}{\partial t} + \frac{\partial j_s^i}{\partial x_i} = q_s \tag{1}$$

Here m_s is the density of a certain physical value (mass, molar amount, energy…); $\boldsymbol{j}_s$ is the flux of this value; q_s is its production, having a meaning of density per time. Thus, for upscaling of Eq. (1), it is necessary to produce the laws for upscaling densities and fluxes. As in the previous work, we do not consider upscaling the field variables (pressure, temperature…) Although both densities and fields belong to the class of intensive values, their upscaling is different (Narasimhan, 1980).

Finally, we do not care much about the mathematical rigorousness of the derivations, assuming e.g. smoothness and differentiability of all the considered variables when necessary.

2. Fundamentals of the continuous upscaling

2.1. Upscaling in one dimension

In this section, we briefly summarize the basic statements of the theory of continuous upscaling of the one-dimensional measures, important for further discussion. For the full account, the reader is addressed to our previous paper (Shapiro, 2021).

Consider a measure $\mu\{\Gamma, t\}$ depending on a set $\Gamma \subseteq \mathbb{R}^1$ and varying with time. Assume that it has a density $m(x, t)$. Typical examples are densities of mass, momentum or energy, mass or molar concentrations, or other values from continuous mechanics.

A transformation of the scale is defined, first, as a transformation of the space and time corresponding to the stretching or contracting them α times: $\Gamma \to \alpha\Gamma$, $t \to \alpha t$. The value of $\alpha > 1$ corresponds to the transition to a finer scale (the set Γ is magnified). It is assumed that time is scaled by the same value of α as the spatial coordinate. Pure scaling $\boldsymbol{S}$ of the measures is simply defined as

$$[\boldsymbol{S}\mu]\{\Gamma, t\} = \mu\left\{\frac{\Gamma}{\alpha}, \frac{t}{\alpha}\right\}; [\boldsymbol{S}m](x, t) = \frac{1}{\alpha} m\left(\frac{x}{\alpha}, \frac{t}{\alpha}\right)$$

Here, for scaling of the density m, the multiplier α^{-1} is used only once because m is the density by x, but not by t.

When considering the three scales, the scaling between the scales 1 and 3 must be a superposition of the scalings between 1 and 2, and 2 and 3: $\alpha_{13} = \alpha_{12}\alpha_{23}$. In this respect, pure scalings form a group.

Apart from scaling operation $\boldsymbol{S}$, we consider the averaging $\boldsymbol{A}$, defined as a linear operation over the measures (or densities) preserving the total measure. Upscaling $\boldsymbol{U}$ is thought of as a superposition of these operations, although this is not postulated from the beginning.

While scaling is possible between any two scales, averaging is defined only when transferring from a finer to a coarser scale. So, the subsequent averagings $\boldsymbol{A}_{ij}$ form a semigroup with regard to their superposition. The same is supposed to be valid for subsequent upscaling operations $\boldsymbol{U}_{ij}$ in general.

Let us introduce a fixed basic scale (0) and number all the other scales by a parameter

$$s = -\ln \alpha, \tag{2}$$

where α is a multiplication coefficient for transfer between the basic and a given scale. An increase of s corresponds to the transition towards the coarser scale. The operators $\boldsymbol{U}_{s_i s_j}$ of transfer between the different scales form a continuous semigroup, in that:

$$\boldsymbol{U}_{s_1 s_3} = \boldsymbol{U}_{s_1 s_2} \boldsymbol{U}_{s_2 s_3} s_1 < s_2 < s_3$$

This makes the operation of continuous upscaling similar to the theory of non-stationary continuous Markov processes. The general Kolmogorov-Feller formalism is fully applicable here (Feller, 1991). It should be adjusted for the task of upscaling, by the introduction of additional limitations. These limitations are related to the physically evident postulate that upscaling of constant and linear densities should result in also constant and linear dependences. It is shown in our previous work (Shapiro, 2021) that upscaling, subject to these limitations, may be described by a specific form of the forward Kolmogorov-Feller equation. Denote by $m_s(x, t)$ the result of upscaling of the initial distribution $m_0(x, t)$ under transition from the reference scale 0 to an arbitrary scale s :

$$m_s(x, t) = \boldsymbol{U}_{0s} m_0(x, t)$$

Then the forward upscaling equation for the density m_s has the form:

$$\frac{\partial m_s}{\partial s} = \boldsymbol{u}_s m_s = \boldsymbol{\tau}_s m_s + \boldsymbol{\xi}_s m_s + \boldsymbol{a}_s m_s; \tag{3}$$

$$\boldsymbol{\tau}_s m_s = t \frac{\partial m_s}{\partial t};$$

$$\boldsymbol{\xi}_s m_s = \frac{\partial}{\partial x}((x - b(s)) m_s);$$

$$\boldsymbol{a}_s m_s = D(s) \frac{\partial^2 m_s}{\partial x^2}$$

Here $\boldsymbol{u}_s$ is the infinitesimal upscaling operator, composed of the operators $\boldsymbol{\tau}_s$, $\boldsymbol{\xi}_s$, and $\boldsymbol{a}_s$. Operators $\boldsymbol{\tau}_s$ and $\boldsymbol{\xi}_s$ describe pure scaling in time and space, correspondingly. They are different, since m_s is a density over space, but not over time. Additionally, operator $\boldsymbol{\xi}_s$ involves an arbitrary displacement $b(s)$ along the x -axis. Operator $\boldsymbol{a}_s$ describes averaging of the density. This averaging, again, happens over space, but not over time; the time scaling is supposed to be exact. Considering the time averaging is also possible but will not be studied here. The expression for $\boldsymbol{a}_s$ is a typical dispersion term, with a diffusion-like coefficient $D(s)$, which may, in principle, depend on the scale s. The fact that $D(s)$ is independent of x, unlike for the general diffusion Markov processes, is a consequence of the assumption that the constant and linear distributions are invariant under upscaling. Eq. (3) means, in particular, that the operations of scaling and averaging are commutative when an "infinitely small" upscaling is considered. For finite upscaling, these operations are not commutative, as shown by Shapiro, 2021.

The scale parameter s plays the role of a new dimension along which the scaling-averaging process is developed. According to Eq. (3), this is a generalized Ornstein-Uhlenbeck process (Feller, 1991; Gardiner, 1985). The properties of this process are well studied. In particular, Eq. (3) allows for an analytical solution, which was studied in the previous paper.

An important general question discussed in Shapiro, 2021, is upscaling of the fluxes. Assume that density m_s obeys a one-dimensional transport equation (conservation law), similar to Eq. (1):

$$\frac{\partial m_s}{\partial t} + \frac{\partial j_s}{\partial x} = q_s$$

Here j_s is the flux, and q_s is a source term (the production density). It was shown that that flux j_s should be upscaled by the same rule as density m_s :

$$\frac{\partial j_s}{\partial s} = \boldsymbol{u}_s j_s = \boldsymbol{\tau}_s j_s + \boldsymbol{\xi} j_s + \boldsymbol{a}_s j_s \tag{4}$$

In particular, Eqs. (3), (4) are valid for an important case of a liquid mass balance equation. In this case, the density m_s is the mass density ρ_s, while the flux j_s is equal to $\rho_s v_s$, where v_s is the convective fluid velocity. A problem is that the expression $\rho_s v_s$ does not only express the mass flux but is also the density of momentum and should automatically be upscaled in correspondence with the rule (3). The proof of Eq. (4) removes a possible disagreement between the two interpretations of this expression, which, in other formalisms, has resulted in a necessity to modify the upscaled momentum equation (Schlechtendahl, 1998).

The previous paper describes also the rules for upscaling of the velocities and non-convective fluxes, which will be considered below for the 3D case.

2.2. The upscaling equation in 3D

The theory of continuous upscaling of the densities allows for a direct generalization onto the three-dimensional case. The densities $m_s(\boldsymbol{x}, t)$ become now functions of time and a point in space $\boldsymbol{x} = (x_1, x_2, x_3) \in \mathbb{R}^3$. For brevity, we will only consider the processes in the three-dimensional space, although the theory may simply be generalized on an arbitrary number of dimensions. We assume that time and all the three dimensions in space are scaled proportionally (cf. Eq. (2)):

$$x_I \rightarrow \alpha x_I = e^{-s} x_I (I = 1, \cdots, 3); t \rightarrow \alpha t = e^{-s} t \tag{5}$$

Likewise, we assume that averaging along the different directions is uniform, with the same dispersion coefficient $D(s)$. Different scalings along the different axes would be a trivial generalization, although the coupled averaging expressed by the dispersion matrix $D_{IK}(s)$ would pose some difficulties. They will not be considered here.

A straightforward generalization of Eq. (3) onto three dimensions is (accounting for our summation agreement, Section 1.2):

$$\frac{\partial m_s}{\partial s} = \boldsymbol{u}_s m_s = \boldsymbol{\tau}_s m_s + \boldsymbol{\xi}_s^i m_s + \boldsymbol{a}_s^i m_s; \tag{6}$$

$$\boldsymbol{\tau}_s m_s = t \frac{\partial m_s}{\partial t};$$

$$\boldsymbol{\xi}_s^I m_s = \frac{\partial}{\partial x_I}((x_I - b_I(s)) m_s)(I = 1, \cdots, 3); \tag{7}$$

$$\boldsymbol{a}_s^I m_s = D(s) \frac{\partial^2 m_s}{\partial x_I^2}(I = 1, \cdots, 3) \tag{8}$$

2.3. Algebra of the transformation operators

For further development, it is convenient to prepare some technical statements, being an extension of Lemmas 5 and 6 from our previous work (Shapiro, 2021). Their proofs are straightforward. The first statement describes the commutation between the different operators entering equation (6) and the operations of differentiation.

Statement 1 (Averaging of the derivatives). Let m be an arbitrary density. Then

$$\boldsymbol{\tau}_s \frac{\partial m}{\partial t} = \frac{\partial}{\partial t}(\boldsymbol{\tau}_s m - m); (\boldsymbol{\xi}_s^I + \boldsymbol{a}_s^I) \frac{\partial m}{\partial t} = \frac{\partial}{\partial t}((\boldsymbol{\xi}_s^I + \boldsymbol{a}_s^I) m);$$

$$\boldsymbol{u}_s \frac{\partial m}{\partial t} = \frac{\partial}{\partial t}(\boldsymbol{u}_s m - m); \left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right) \frac{\partial m}{\partial t} = \frac{\partial}{\partial t}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right) m + m\right)$$

Similarly,

$$\boldsymbol{\tau}_s \frac{\partial m}{\partial x_K} = \frac{\partial}{\partial x_K}(\boldsymbol{\tau}_s m); (\boldsymbol{\xi}_s^I + \boldsymbol{a}_s^I) \frac{\partial m}{\partial x_K} = \frac{\partial}{\partial x_K}((\boldsymbol{\xi}_s^I + \boldsymbol{a}_s^I) m - \delta_{IK} m);$$

$$\boldsymbol{u}_s \frac{\partial m}{\partial x_K} = \frac{\partial}{\partial x_K}(\boldsymbol{u}_s m - m); \left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)\frac{\partial m}{\partial x_K} = \frac{\partial}{\partial x_K}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)m + m\right)$$

Statement 2 (Averaging of a product). Let m, n be arbitrary densities. Then

$$\boldsymbol{u}_s(mn) = m\boldsymbol{u}_s n + n\boldsymbol{u}_s m - \alpha mn + 2D_0(s)\frac{\partial m}{\partial x_i}\frac{\partial n}{\partial x_i};$$

$$\left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)(mn) = m\left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)n + n\left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)m + \alpha mn - 2D_0(s)\frac{\partial m}{\partial x_i}\frac{\partial n}{\partial x_i} \tag{9}$$

Coefficient α in these equations depends on a dimension of the space. In 1D, it is equal to 1. If the 1D-densities m, n depend also on time, it becomes 2. Similarly, α is equal to 3 for stationary (time-independent) 3D densities, and 4 for time-dependent densities in 3D.

2.4. Upscaling of the fluxes

It was demonstrated in the previous work (Shapiro, 2021) that the flux in one dimension obeys the same equation (4) as the density. In three dimensions, this statement is not valid anymore. Its generalization is considered in this section.

Consider an arbitrary balance/flow equation (1). We assume that this equation should be valid for any scale, which should be secured by an upscaling rule for the flux $\boldsymbol{j}_s$. The source (production) term q_s is thought of as a change of some density n_s per unit time: $q_s = \partial n_s/\partial t$. Hence scaling of q_s should obey the rule formulated in Statement 1:

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)q_s = \frac{\partial}{\partial t}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)n_s + n_s\right) = \frac{\partial n_s}{\partial t}$$

The last equality is valid, since n_s is density obeying Eq. (6). Finally,

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)q_s = q_s \tag{10}$$

After this preparation, we apply operator $\frac{\partial}{\partial s} - \boldsymbol{u}_s$ to Eq. (1). Accounting for Eq. (10), we obtain:

$$\left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)\frac{\partial m_s}{\partial t} + \left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)\frac{\partial j_s^i}{\partial x_i} = q_s \#$$

Interchanging the derivatives with the operators according to Statement 1, we obtain that

$$\frac{\partial}{\partial t}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)m_s + m_s\right) + \frac{\partial}{\partial x_i}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)j_s^i + j_s^i\right) = q_s$$

In this equation, the term $\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)m_s$ is eliminated, since this is the upscaling equation for density. The terms $\frac{\partial m_s}{\partial t}$ from the first and $\frac{\partial j_s^i}{\partial x_i}$ from the second addendum are combined with q_s on the right-hand side and canceled due to Eq. (1). The resulting equality is

$$\frac{\partial}{\partial x_i}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)j_s^i\right) = 0 \tag{11}$$

Vice versa, assume that Eq. (11) is valid, and denote $\frac{\partial m_s}{\partial t} + \frac{\partial j_s^i}{\partial x_i}$ by q_s. Then, the inverse substitution may be performed, starting from Eq. (11) and resulting in Eq. (10). Thus, q_s is really upscaled as a production term, and Eq. (1) remains valid for any scale.

A particularly important case is the case of zero production: The quantity m_s is conserved, so that the production $q_s = 0$. It is easy to see from Eq. (10) that, if $q_s = 0$ for some scale, then it remains zero for any scale. In this case, Eq. (11) guarantees that the conservation law (1) with $q_s = 0$ is preserved under upscaling. This equation is a necessary and sufficient condition for the preservation of the conservation law. Indeed, assume that Eq. (11) is valid. Since m_s is a

density, it obeys the upscaling equation (6), and Eq. (11) may be extended to

$$\frac{\partial}{\partial t}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)m_s\right) + \frac{\partial}{\partial x_i}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)j_s^i\right) = 0$$

Exchanging the derivatives with the operator $\frac{\partial}{\partial s} - \boldsymbol{u}_s$ according to statement 1, we obtain:

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)\left(\frac{\partial m_s}{\partial t} + \frac{\partial j_s^i}{\partial x_i}\right) - \left(\frac{\partial m_s}{\partial t} + \frac{\partial j_s^i}{\partial x_i}\right) = 0$$

Thus, application of the evolution operator $\frac{\partial}{\partial s} - \boldsymbol{u}_s - \boldsymbol{I}$ to the expression $\frac{\partial m_s}{\partial t} + \frac{\partial j_s^i}{\partial x_i}$ results in zero. If this expression is equal to zero at a certain scale, it will be zero at any other scale.

In 1D, Eq. (11) may be integrated over x, resulting in Eq. (4) for j_s. This is not true in multiple dimensions, where a single equation (11) is insufficient for determining all the components of the upscaled flux. Additional considerations are required to determine the upscaling rules for the flux components.

As an example, consider the mass flux $\boldsymbol{j} = \rho\boldsymbol{v}$. This expression has a dual character, as was discussed in the previous work (Shapiro, 2021). On one hand, this is the expression for the flux in the mass conservation law; on the other hand, this is the density of momentum. Hence, its components should be upscaled like any other density:

$$\left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)\rho_s v_s^I = 0 (I = 1, \cdots, 3) \tag{12}$$

This rule does not contradict Eq. (11), but it is more definite. Thus, in this case, a particular form of the flux makes it possible to formulate a rule for upscaling of each of its components. Further, application of Statement 2 (Eq. (9)) to Eq. (12) and utilizing the fact that density ρ_s is canceled by the operator $\frac{\partial}{\partial s} - \boldsymbol{u}_s$ results in the upscaling equation for the components of the velocity:

$$\left(\tfrac{\partial}{\partial s} - \boldsymbol{u}_s\right)v_s^I = -\alpha v_s^I + 2D_0(s)\frac{\partial \ln \rho_s}{\partial x_k}\frac{\partial v_s^I}{\partial x_k} \tag{13}$$

Consider now upscaling of the non-convective fluxes. A general flow equation (1) is often expressed in the form

$$\frac{\partial m_s}{\partial t} + \frac{\partial(v_s^i l_s + L_s^i)}{\partial x_i} = q_s$$

Here $\boldsymbol{L}_s$ is the non-convective flux, and l_s is the transfer density, which may be different from m_s (for example, for the energy transfer in liquids m_s is the energy density, while l_s is the specific enthalpy). Eq. (11) for this case becomes

$$\frac{\partial}{\partial x_i}\left(\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)\left(v_s^i l_s + L_s^i\right)\right) = 0$$

Subsequent application of Statement 2 and rules (6) and (13) results in

$$\frac{\partial}{\partial x_i}\left[\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)L_s^i - 2D_0(s)\frac{\partial v_s^i}{\partial x_k}\left(\frac{\partial l_s}{\partial x_k} - \frac{l_s}{m_s}\frac{\partial m_s}{\partial x_k}\right)\right] = 0 \tag{14}$$

In the two important cases, this rule for upscaling the non-convective flux $\boldsymbol{L}_s$ is the same as the rule (11) for upscaling the convective flux. The term $2D_0(s)\frac{\partial v_s^i}{\partial x_k}\left(\frac{\partial l_s}{\partial x_k} - \frac{l_s}{m_s}\frac{\partial m_s}{\partial x_k}\right)$ in Eq. (14) is canceled if the convective flux is absent: $\boldsymbol{v}_s = 0$. Another case is where the density m_s is equal to the transfer density l_s. This is true, for example, for the momentum transfer, where both densities are equal to $\rho_s\boldsymbol{v}_s$. The equality $m_s = l_s$ is also true for the diffusion of an additive. In this case, both m_s and l_s are equal to the product $\rho_s c_s$ of the density and the concentration of the additive.

In sum, conditions (11) and (14) are necessary for upscaling of the fluxes, but they do not provide a sufficient rule for their upscaling. Unlike the 1D case, additional assumptions about the structure of the fluxes are required. For example, for the process of diffusion,

the proportionality of the non-convective diffusion flux L_s to the gradient of concentration may be utilized. This program is carried out in the next section.

3. Diffusion in 3D

3.1. Statement of the upscaling problem

The simplest form of the traditional 3D steady-state diffusion equation in a heterogeneous medium is

$$\frac{\partial}{\partial x_i}\left(d(\boldsymbol{x})\frac{\partial c}{\partial x_i}\right) = 0 \tag{15}$$

Here $c(\boldsymbol{x})$ is the particle concentration; $d(\boldsymbol{x})$ is the diffusion coefficient. Eq. (15) corresponds to a constant density ρ of the solution. The assumption about the constancy of the density is not restrictive: otherwise, the diffusion coefficient in the following derivations may be substituted by ρd.

A fundamental problem about the diffusion equation is, whether upscaling retains its form, or additional terms are needed. This problem is nontrivial, as discussed by Quintard and Whitaker, 1994a,b,c; Panfilov and Rasoulzadeh, 2013; Rasoulzadeh et al., 2014; see also references therein. In many studies, upscaling is carried out, implicitly or explicitly, for the *particular* solutions c of Eq. (15), and it is not discussed whether the resulting upscaling equation is valid for *all* the solutions. Reformulating the problem, it may be asked, for what *class* of the solutions c Eq. (15) remains valid for all the scales, and how should the diffusion coefficient be correspondingly upscaled.

Consider, for example, a single solution c of Eq. (15). If only this solution is of interest, then upscaling of the diffusion coefficient is possible, as follows from the following lemma:

Lemma 1.. *For any smooth non-degenerate dependence $c(\boldsymbol{x})$, such a positive function $d(\boldsymbol{x})$ exists that Eq. (15) is valid.*

Proof. Eq. (15) may be rewritten in the form

$$\frac{\partial c}{\partial x_i}\frac{\partial \ln d}{\partial x_i} = -\frac{\partial^2 c}{\partial x_j^2} \tag{16}$$

Under known c, this is a first-order linear hyperbolic equation for $\ln d$. It may be solved provided that the boundary condition for $\ln d$ is specified on a surface, which is not tangent to ∇c. The exponent of the solution gives a positive value of d.

Upon validity of Lemma 1, the algorithm for upscaling the diffusion coefficient with a single solution may be outlined as follows. Assume that the diffusion coefficient $d(\boldsymbol{x})$ and the corresponding concentration $c(\boldsymbol{x})$ are known at some scale zero. Since c is a density, its upscaling may be carried out by solving Eq. (6), and the set of upscaled concentrations $c_s(\boldsymbol{x})$ may thus be determined independently of the diffusion coefficient. Then, for each c_s, the corresponding coefficient $d_s(\boldsymbol{x})$ may be found from Eq. (16). It serves as an upscaled coefficient of d to the scale s, preserving the form of the diffusion equation.

The described "upscaling procedure" has several obvious shortcomings. First, the diffusion coefficient $d_s(\boldsymbol{x})$ depends on a particular density c. It cannot be guaranteed that taking another solution would not result in a different coefficient d_s. The upscaling procedure is thus not universal. Secondly, the upscaling is not unique. It depends on a boundary condition for Eq. (16), which may be different for the different scales. Finally, the coefficients d_s for each scale are determined individually, without considering other scales. They are disconnected from each other.

As shown by Shapiro, 2021 (and reproduced below, with some modifications and extensions), a single universal equation for upscaling of d_s, regardless of the solution c_s, exists in 1D case. How-

ever, this is not true in multiple dimensions. The proof will be given later on (Section 3.3).

In what follows in this section, we consider a generalization of the 1D upscaling equation for the diffusion coefficient, onto three dimensions.

3.2. Upscaling of the diffusion coefficient in 1D

3.2.1. The diffusion equation without the source terms

Before considering the 3D case, let us derive the upscaling equation for the diffusion coefficient in 1D, generalizing the derivation by Shapiro, 2021 (also, a small error is corrected).

Consider Eq. (11) in one spatial dimension ($x_1 = x; j_s^1 = j_s$). Its integration over x formally results in

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)j_s = E(t)$$

Since upscaling of the zero flux should result in zero j_s, the only possibility is $E(t) = 0$. Hence, the flux in 1D is upscaled by the same rule as the density. Moreover, as discussed after Eq. (14), for the diffusion process the non-convective flux is upscaled by the same rule as the convective flux:

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)L_s = 0 \#$$

If the liquid density is constant, the diffusion flux is proportional to the gradient of the concentration:

$$j_s = d_s(x)\frac{\partial c_s}{\partial x}$$

Hence, the upscaling equation assumes the form of

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)\left(d_s(x)\frac{\partial c_s}{\partial x}\right) = 0$$

Subsequent application of statement 2 to the product; then, of statement 1 to the derivative $\frac{\partial c_s}{\partial x}$; and, finally, noticing that c_s is canceled by the operator $\frac{\partial}{\partial s} - \boldsymbol{u}_s$, results in

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)\left(d_s(x)\frac{\partial c_s}{\partial x}\right) = \frac{\partial c_s}{\partial x}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s(x) + (\alpha+1)d_s(x)\frac{\partial c_s}{\partial x} - 2D_0(s)\frac{\partial d_s}{\partial x}\frac{\partial^2 c_s}{\partial x^2} \tag{17}$$

In 1D, we have $\alpha = 1$. From Eq. (15) in one dimension, it follows that

$$\frac{\partial^2 c_s}{\partial x^2} = -\frac{1}{d_s(x)}\frac{\partial d_s}{\partial x}\frac{\partial c_s}{\partial x} \tag{18}$$

Substitution of this equation to Eq. (17) results in

$$\frac{\partial c_s}{\partial x}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s(x) + 2d_s(x)\frac{\partial c_s}{\partial x} + 2D_0(s)\frac{1}{d_s(x)}\left(\frac{\partial d_s}{\partial x}\right)^2\frac{\partial c_s}{\partial x} = 0$$

The multiplier $\frac{\partial c_s}{\partial x}$ is common and may be reduced. The resulting equation for the diffusion coefficient becomes

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s(x) + 2d_s(x) + 2D_0(s)\frac{1}{d_s(x)}\left(\frac{\partial d_s}{\partial x}\right)^2 = 0 \tag{19}$$

This is a nonlinear equation, which will be investigated numerically later. It is important to notice that this equation is independent of a solution for the concentration and makes it possible to upscale the diffusion coefficient independently.

The described derivation of the upscaling equation for the diffusion coefficient has several limitations. First, it is only valid for a steady state. For a non-stationary diffusion, $\frac{\partial^2 c_s}{\partial x^2}$ cannot be expressed in terms of $\frac{\partial c_s}{\partial x}$, and Eq. (17) cannot be further simplified.

Further, the suggested procedure has no meaning for an important case where the diffusion coefficient is equal to zero in some regions. This is the case of diffusion in a porous medium, where diffusion through the solid is impossible. If in one dimension some

regions, or even points, are excluded from diffusion, then the regions where diffusion occurs become disconnected, and diffusion through the whole space becomes impossible. In multiple dimensions, this is not the problem since the free space may be multiply connected. The diffusing particles may bypass the obstacles by moving around them.

Formally, this problem of impermeable inclusions may be avoided by assuming that the diffusion coefficient is equal to a very small value where it should be zero. This is a necessary simplification for a numerical solution of Eq. (19), and of the similar 3D equations derived later since they involve division by $d_s(x)$. This trick will be exploited in the numerical simulations.

3.2.2. The 1D diffusion with a source.

It may be tempting to claim that the equation (19) derived in the previous section is as universal as Eq. (6) for upscaling the densities or Eq. (11) for upscaling the fluxes. Unfortunately, this is not the case. Consider the stationary diffusion equation with the source term:

$$\frac{\partial}{\partial x}\left(d_s(x)\frac{\partial c_s}{\partial x}\right) = q_s(x) \tag{20}$$

The derivation from the previous section remains valid until the equation (18), which is modified:

$$\frac{\partial^2 c_s}{\partial x^2} = \frac{q_s(x)}{d_s(x)} - \frac{1}{d_s(x)}\frac{\partial d_s}{\partial x}\frac{\partial c_s}{\partial x}$$

Further repetition of the steps from the previous derivation ends with:

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s(x) + 2d_s(x) + 2D_0(s)\frac{1}{d_s(x)}\left(\frac{\partial d_s}{\partial x}\right)^2 - 2D_0(s)\frac{\partial d_s}{\partial x}\frac{q_s(x)}{d_s(x)\frac{\partial c_s}{\partial x}} = 0\#$$

For exclusion of the concentration c_s from the last addendum, Eq. (20) is applied again. It is integrated over x to express $d_s(x)\frac{\partial c_s}{\partial x}$. Substitution results in:

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s(x) + 2d_s(x) + 2D_0(s)\frac{1}{d_s(x)}\left(\frac{\partial d_s}{\partial x}\right)^2 - 2D_0(s)\frac{\partial d_s}{\partial x}\frac{q_s(x)}{\int q_s(x)dx} = 0 \tag{21}$$

This equation differs from Eq. (19) by the last term depending on the source q_s. The upscaling equation for the diffusion coefficient is thus dependent on the source term in the equation. Even less trivially, if q_s tends to zero, the corresponding term in the upscaling equation (21) does not disappear. As an example, consider the source q_s independent of x. It follows from Eq. (10) that in this case the value of q_s is equal to $q_0 e^{2s}$. Substitution into Eq. (21) results in

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s(x) + 2d_s(x) + 2D_0(s)\frac{1}{d_s(x)}\left(\frac{\partial d_s}{\partial x}\right)^2 - 2D_0(s)\frac{\partial d_s}{\partial x}\frac{1}{x - x_0}$$
$$= 0$$

Here x_0 is the integration constant, to be found from the initial condition. The value of q_0 does not appear in the last equation. If this value tends to zero, the last term does not disappear and, contrary to expectations, the upscaling equation (21) is not reduced to Eq. (19).

Independence of the upscaling equation on the size of the source q_s may be explained by the fact that, if the source term in the diffusion equation (20) is rescaled, the concentration c_s should be rescaled correspondingly, while the diffusion coefficient remains unchanged. The value $q_s = 0$ is a singular point.

Although the upscaling equation (21) is independent of the size of q_s, it still depends on the shape of the function $q_s(x)$. In particular, in the regions where $q_s(x) = 0$, Eq. (qs1Dq) is reduced to Eq. (qs1D).

One of the long-discussed and contradictory problems of the reactive flows in porous media is, whether the effective macroscale diffusivity in a catalyst depends on the reaction rate. See the reviews of the history of the problem in Valdés-Parada et al., 2011; 2017. Although the considered problem is not equivalent to the diffusion with chemical reactions, there is an analogy between them, since the right-hand side of the equation may, to some extent, be interpreted as a source of concentration produced by the reaction. In this respect, the considered upscaling scheme confirms the dependence of the upscaled diffusion coefficient on the reaction rate. A more detailed analysis may be subject to a separate study.

3.3. Upscaling diffusion in three dimensions

3.3.1. The general equation for upscaling

In 3D, the components of the non-convective diffusion flux are expressed as

$$L_s^K = d(\boldsymbol{x})\frac{\partial c_s}{\partial x_K}\,(K = 1, \cdots, 3)$$

We apply the operator $\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)$ to these expressions. Applying Statement 2 to the product and, subsequently, Statement 1 to the derivative $\frac{\partial c_s}{\partial x_K}$, we obtain

$$\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)L_s^K = \frac{\partial c_s}{\partial x_K}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s + 4d_s\frac{\partial c_s}{\partial x_K} - 2D_0(s)\frac{\partial d_s}{\partial x_i}\frac{\partial}{\partial x_i}\frac{\partial c_s}{\partial x_K}$$

This expression is substituted to Eq. (14) (without the term arising from convection). The second term is then canceled due to Eq. (15). The resulting equation has the form of

$$\frac{\partial}{\partial x_k}\left[\frac{\partial c_s}{\partial x_k}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s - 2D_0(s)\frac{\partial d_s}{\partial x_i}\frac{\partial^2 c_s}{\partial x_i\partial x_k}\right] = 0 \tag{22}$$

Unlike in the 1D case, it is impossible to integrate this equation and to separate the diffusion coefficient from the concentration. In the following subsections, we will consider important particular cases where the derivation of a single equation for upscaling the diffusion coefficient becomes possible. By comparison of these cases, we will show that the different equations for d_s may be valid depending on, what solution for c_s is chosen.

3.3.2. The factorized diffusion coefficient

Consider a model medium as shown in Fig. 1 (in 2D projection) and often used as an example in the theory of upscaling (Ryan et al., 1980; Arbogast et al., 1990). Similar models are studied in the theory of homogenization of the flows in fractured porous media (Panfilov and Rasoulzadeh, 2013). The periodic model consists of cubic "blocks" or "catalyst pellets" with a low diffusion coefficient, surrounded by "fractures", or "channels" between them, where the diffusion coefficient is high. The distribution of the diffusion coefficients for such a system may be approximated by

$$d(\boldsymbol{x}) = d^1(x_1)d^2(x_2)d^3(x_3), \tag{23}$$

where the coefficients $d_I(x_I)$ are expressed in terms of the Heaviside function H:

$$d^I(x_I) = \sqrt[3]{d_0}\left[\varepsilon^{\frac{1}{3}}\sum_{k=-\infty}^{\infty}\left(H\left(x_I - Y_I^k\right) - H\left(x_I - Z_I^k\right)\right) + \varepsilon^{-\frac{2}{3}}\sum_{k=-\infty}^{\infty}\left(H\left(x_I - Z_I^k\right) - H\left(x_I - Y_I^k\right)\right)\right]; \tag{24}$$

$$Y_I^K = \frac{a}{2} + (K-1)(a+b); Y_I^K = \frac{a}{2} + (K-1)a + Kb$$

Here a is the width of the channel, and b is the cube size. With such coefficients d^I, the diffusion coefficient will be equal to $d_0\varepsilon$ in the blocks, d_0 in the channels, $d_0\varepsilon^{-1}$ in the intersections of two
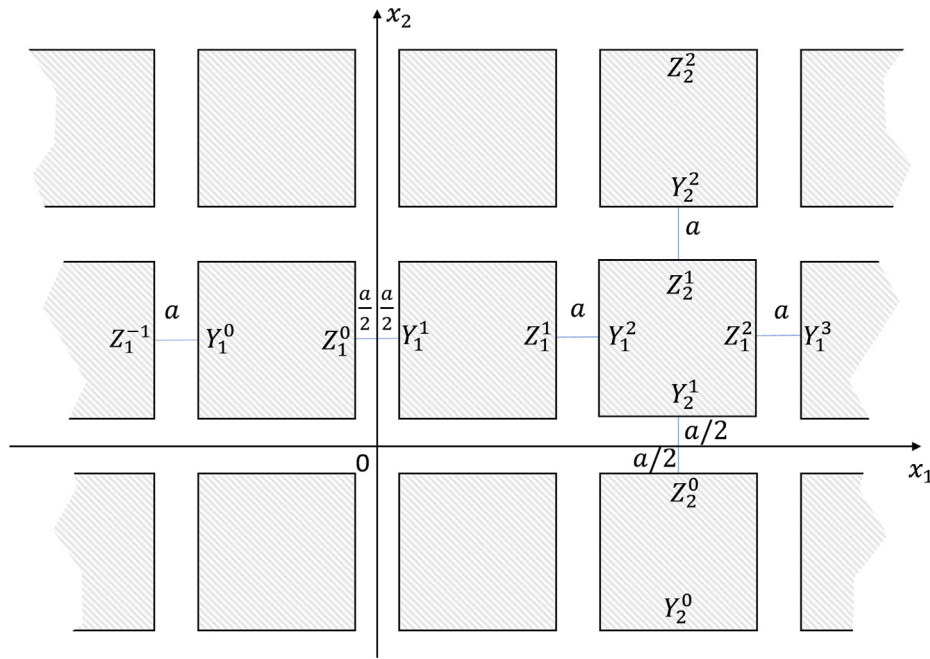
Fig. 1. A periodic structure of high-permeable channels and low-permeable blocks. In another interpretation, the structure consisting of high-permeable cavities and low-permeable borders.

channels, and $d_0 \varepsilon^{-2}$ in the intersections of three channels. Obviously, for small ε, the diffusion coefficient in the blocks is much lower than in the channels. If the ratio a/b is small, the role of the channel intersections in total diffusion is negligible, and the model serves a good approximation for the blocks and channels with uniform diffusion coefficients. Thus, formula (24) may be used as a simple model for the diffusion in a medium with high-permeable channels and low-permeable inclusions. This model is also more convenient for the mathematical analysis than the traditional model with uniform channels, since (as we show below) it allows for the analytical solution.

Similarly, the factorized diffusion coefficient (23) may be applied to model the flows in the media with high-conductivity inclusions. It may be set, for example,

$$d^I(x_I) = \sqrt[3]{d_0}\left[\varepsilon^{-\frac{1}{3}}\sum_{k=-\infty}^{\infty}\left(H\left(x_I - Y_I^k\right) - H\left(x_I - Z_I^k\right)\right) + \varepsilon^{\frac{2}{3}}\sum_{k=-\infty}^{\infty}\left(H\left(x_I - Z_I^k\right) - H\left(x_I - Y_I^k\right)\right)\right]; \#$$

(25)

These examples illustrate the practical importance of diffusion in heterogeneous media with the factorized diffusion coefficients. We will consider a general problem where the diffusion coefficient may be factorized according to Eq. (23), but not necessarily in the forms (24) or (25) (the coefficients d^I do not even have to be periodic).

The diffusion equation (15) with a factorized coefficient allows for multiple solutions. One class of such solutions is where the concentration is also factorized:

$$c(\boldsymbol{x}) = c^1(x_1)c^2(x_2)c^3(x_3),$$

(26)

The factors c^I may be selected to obey individual "diffusion" equations:

$$\frac{\partial}{\partial x_I}\left(d^I(x_I)\frac{\partial c^I}{\partial x_I}\right) = 0$$

(27)

A more general form of c^I obeys the non-homogeneous equations with arbitrary constants H_I:

$$\frac{1}{d^I c^I}\frac{\partial}{\partial x_I}\left(d^I(x_I)\frac{\partial c^I}{\partial x_I}\right) = H_I, \sum_I H_I = 0$$

(28)

It should be stressed that the factorized concentrations (26) are only particular, but not general solutions of the diffusion equation with the factorized coefficient (23). For example, not all the solutions of the diffusion equation with a constant diffusion coefficient are factorized. The inverse statement is not valid, either. As shown in Appendix A, the most general form of the diffusion coefficient corresponding to the concentration (26) may contain a non-factorized multiplier.

Another class of the solutions for the diffusion equation with the factorized coefficient (26) is:

$$c(\boldsymbol{x}) = c^1(x_1) + c^2(x_2) + c^3(x_3),$$

(29)

Such a solution obeys the diffusion equation (15) if the addenda obey the equation (27). More generally, they may obey the equations, slightly different from (28):

$$\frac{1}{d^I}\frac{\partial}{\partial x_I}\left(d^I(x_I)\frac{\partial c^I}{\partial x_I}\right) = H_I, \sum_I H_I = 0$$

(30)

By application of the upscaling equation (6) to the factorized concentrations in the forms (26) or (29), it may be established that the upscaled concentrations are also factorized:

$$c_s(\boldsymbol{x}) = c_s^1(x_1)c_s^2(x_2)c_s^3(x_3), \text{or} c_s(\boldsymbol{x}) = c_s^1(x_1) + c_s^2(x_2) + c_s^3(x_3)\#$$

In both cases, the individual concentrations c_s^I may be selected to be the densities of some measures along the corresponding coordinate x^I, that is, to obey the equations (cf. Eqs. (7), (8)):

$$\frac{\partial c_s^I}{\partial s} = \boldsymbol{u}_s^I c_s^I = \xi_s^I c_s^I + \boldsymbol{a}_s^I c_s^I$$

(31)

In the following, we will derive the upscaling equations for the factorized diffusion coefficient (23), for the following three cases:

Case 1.. *Concentration c_s is factorized into a product (26) of concentrations $c_s^I(x_I)$, each obeying the equations (27).*

Case 2.. *Concentration c_s is represented as a sum (29) of concentrations $c_s^I(x_I)$, each obeying the equations (27).*

Case 3.. *Concentration c_s is represented as a sum* (29) *of concentrations* $c_s^l(x_l)$, *each obeying the diffusion equations* (30), *for arbitrary constants* H^l.

We have also considered case 4, where the concentration is factorized into the product (26), and each c^l obeys Eq. (28). Unfortunately, for this case, it does not seem to be possible to derive an upscaling equation for the diffusion coefficient. The same is true about the case where the diffusion coefficient is represented as a sum of $d^i(x_i)$. For this case, the solutions in the form of (26) or (29) also exist, and the concentrations $c^i(x_i)$ may be upscaled individually. However, the transformations like those presented below do not work, and separate upscaling equations for the diffusion coefficient cannot be derived.

3.3.3. Upscaling for case 1

The procedure of derivation of the equations for the diffusion coefficient factors d^K is cumbersome and not informative. The derivation is outlined in Appendix B. The resulting equation is:

$$\frac{\partial}{\partial x_K}\left\{\frac{1}{d_s^K}\left(\frac{\partial}{\partial s}-\boldsymbol{u}_s^K\right)d_s^K + 2D_0(s)\frac{1}{\left(d_s^K\right)^2}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2\right\}=0\,(K=1,\cdots 3) \qquad (32)$$

Eq. (32) is formally different from Eq. (19) for the one-dimensional diffusion coefficient. It may be transformed to a similar form by integration over x_K and multiplication by d_s^K:

$$\left(\frac{\partial}{\partial s}-\boldsymbol{u}_s^K\right)d_s^K + \beta(s)d_s^K + 2D_0(s)\frac{1}{d_s^K}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2 = 0 \qquad (33)$$

Here $\beta(s)$ is the constant of integration. The last equation is reduced to Eq. (19) for the 1D case if this constant is set to be equal to 2. For the 3D case, this constant must be determined separately.

To determine $\beta(s)$ for the 3D diffusion, the case of a constant coefficient d_s may be considered. It follows from Eq. (6) that this coefficient will be independent of $\boldsymbol{x}$ if the initial, non-upscaled coefficient d_0 is constant. The dimensionality of the diffusion coefficient is L^2/T (distance squared by time). Then it should be rescaled according to Eq. (5):

$$d_s = \frac{\alpha^2}{\alpha}d_0 = e^{-s}d_0 \qquad (34)$$

Upon factorization, it is reasonable (though not necessary) to assume that the coefficients d_s^K are scaled equally. Then

$$d_s^K = e^{-\frac{s}{3}}d_0^{\frac{1}{3}}$$

Substitution of these coefficients into Eq. (33) (note the special role of ξ_s^K) results, after transformations, in the value of $\beta = \frac{4}{3}$. In a more general multidimensional case, β is equal to $1 + D^{-1}$, where D is the dimension of the space. For calculations, it is important to know that β is a constant independent of s.

Eqs. (33) or (32) provide the rules for upscaling of the individual multipliers in the factorized diffusion coefficient. A question may arise, whether it is possible to produce an upscaling equation for the whole coefficient d_s, "forgetting" that it is factorized. An additional requirement may be that this equation is symmetric with regard to the different spatial coordinates. An answer may be obtained by multiplying each of the equations (33) by all $d_s^i, i\neq K$, and adding them together. The final equation is

$$\left(3\frac{\partial}{\partial s}-\boldsymbol{u}_s\right)d_s + 4d_s + \frac{1}{d_s}\left(\frac{\partial d_s}{\partial x_i}\right)^2 = 0 \qquad (35)$$

This equation differs from the 1D equation (19) by the coefficients at $\frac{\partial}{\partial s}$ and at the second addendum. Both coefficients are related to the dimension of the space.

3.3.4. Cases 2 and 3

Wonderfully, case 2 (the additive concentration (29)) results in the same upscaling equations for the diffusion coefficient, (32) to (35), as derived in the previous subsection. The derivation is not presented here. It is similar to the derivation of Appendix B, with a correction for the new differentiation rules (46) formulated in Appendix C.

The fact that the upscaling equations for Cases 1 and 2 are the same might raise hopes that the upscaling equation (35) is universal. Unfortunately, this is not true. Derivations of Appendix C show that in case 3 the upscaling equations for coefficients d_s^l may be qualitatively different from Eq. (32) and its consequence (33). If the individual concentrations in the decomposition (29) are selected to obey Eq. (30), the upscaling equation for d_s^l becomes (47):

$$\frac{\partial}{\partial x_K}\left[\frac{1}{d_s^K}\left(\frac{\partial}{\partial s}-\boldsymbol{u}_s^K\right)d_s^K + 2D_0(s)\frac{1}{\left(d_s^K\right)^2}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2\right] + 2D_0(s)\frac{1}{\int d_s^K dx_K}\left[\frac{1}{d_s^K}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2 - \frac{\partial^2 d_s^K}{\partial x_K^2}\right]=0\# \qquad (36)$$

A remarkable property of this equation is that it is independent of the constants H^K. The remarkableness of this property becomes clear when considering the constants H^K tending to zero: then Eq. (36) does not converge to a simpler equation (32), as might be expected. Independence of constants H^K is similar to such property of the 1D diffusion equation with the right-hand side described in Section 3.2.2, and Eq. (36) is, to some extent, similar to Eq. (21). However, there is an important difference. Eq. (21) is different from the upscaling equation (19) because the former equation is related to the diffusion equation with a source, while the latter is valid for the equation without the source of concentration. The difference between them may be attributed to the different physical processes that they describe. Meanwhile, Eq. (32) and Eq. (36) are the upscaling equations for the same 3D diffusion equation without the source of concentration. They correspond to the different solutions of this equation. Thus, we have proven that upscaling of the diffusion coefficient may be different for the different solutions of the same diffusion equation.

Similar to the previous section (Eq. (35)), it is possible to unite the equations for separate coefficients d_s^K and produce a single equation for the diffusion coefficient d_s:

$$\frac{\partial}{\partial x_k}\left[\frac{1}{d_s}\left(3\frac{\partial}{\partial s}-\boldsymbol{u}_s\right)d_s + 2D_0(s)\frac{1}{(d_s)^2}\left(\frac{\partial d_s}{\partial x_k}\right)^2\right] + 2D_0(s)\frac{1}{\int d_s dx_k}\left[\frac{1}{d_s}\left(\frac{\partial d_s}{\partial x_k}\right)^2 - \frac{\partial^2 d_s}{\partial x_k^2}\right]=0 \qquad (37)$$

The structure of this equation is much more complex than of Eq. (32). It may be reduced to the former equation only if the last term may be neglected. Then the rest equation may be integrated by x_k, resulting in Eq. (32). A question arises, whether both equation converge to each other and produce the same value of the diffusion coefficient asymptotically, as $s \to \infty$. If demonstrated, this property would provide a partial answer to the question, whether the asymptotic averaged value of the diffusion coefficient is independent of the solution for the concentration, similar to other homogenization theories. An outline of the proof may be as follows. Equations (35) and (37) should produce close results at large values of x_k, where the integral in the denominator of the last term in Eq. (37) becomes large. Rescaling results in bringing the areas of large values of x_k closer to zero, so that the interval where the whole term is significant shrinks. However, this outline is still far from a finished proof. The problem needs a separate study.

3.3.5. Discussion

Can the diffusion coefficients be upscaled without any knowledge about a solution for the diffusion equation? This study gives a double answer to this question.

For the diffusion without sources in 1D, the answer is positive. Eq. (19) may be used for upscaling the diffusion coefficient. A similar equation (35) may be used in three dimensions. Unlike in a single dimension, this equation is not universally valid but is only applicable to a rather specific statement described in Section 3.3.2. However, this statement may be sufficient to model several practically important situations.

On the other hand, the rules for upscaling of the diffusion coefficient in 1D become different if the diffusion equation with sources is considered. This difference is not reduced if the source term tends to zero. Thus, even in 1D, the answer given by Eq. (19) is not universal. A more complex upscaling equation (21) should be applied to upscale the diffusion coefficient for the diffusion with a source.

In three dimensions, the situation is even more complicated. For some classes of the solutions of the diffusion equation, the upscaling equation for the diffusion coefficient becomes rather complex, assuming the form (37). For other solutions or other forms of factorization of the diffusion coefficient, upscaling may become impossible. The general equation (22) should be applied for upscaling of the diffusion coefficient, linked to the solution for the concentration c_s found by application of the upscaling equation (6). This is necessary, for example, if it is important to account for the effect of bypassing the low-permeable obstacles by going around them. The factorized solutions for concentration, (26) and (29), do not describe this effect.

In the previous derivations, it was assumed that the diffusion equation retains its form (15) under upscaling. This assumption seems natural for continuous upscaling; however, it was not always kept in other upscaling methods. Apart from the appearance of the higher derivatives (Quintard and Whitaker, 1994a,b,c; Whitaker, 1999) and nonlocal equations (Panfilov and Rasoulzadeh, 2013; Rasoulzadeh et al., 2014), some authors proposed the two-continuum model, where the flow is modeled separately in the two overlapping continua of high-permeable channels and low-permeable blocks, and the exchange between them occurs (Barenblatt et al., 1960; Bedrikovetsky and Rowan, 1993; Lim and Aziz, 1995). The works of Ahmadi et al., 1998; Golfier et al., 2007 contain the substantiation of this approach for diffusion in the framework of the volume averaging, and its comparison with the available experimental data. It is concluded that the two-continuum model may catch most of the peculiarities of the flow; however, still, there are some features of the model (early arrival of the concentration front, massive tails) that are not in full agreement with the experiments. Other approaches may be more efficient there, like the continuous-time random walks (CTRW) (Montroll and West, 1979; Berkowitz et al., 2006; Shapiro and Bedrikovetsky, 2008; Shapiro and Yuan, 2011). Their comparison is outside of the scope of the present work.

3.4. Numerical solution

3.4.1. Statement of the problem and the method for the solution

We have solved the upscaling equation numerically, to demonstrate its properties and study the behavior of the solution. This study concerns only basic features; the detailed analysis and comparison to multiple previous works require a separate investigation. We have solved the 1D homogeneous upscaling equation (19). The 3D equations (33), (35) are similar, while Eq. (36) is much more complicated and will not be considered here.

For the numerical solution, it is convenient to transfer to the logarithm of the diffusion coefficient. This makes it possible to avoid divisions by $d_s(x)$ and to regularize the computations where the diffusion coefficient approaches zero. Further, we substitute

$$x = e^{-s}y \qquad (38)$$

Denoting $E_s(y) = \ln d_s(e^{-s}y)$, we represent the equation in the form of

$$\frac{\partial E_s}{\partial s}\Big|_y = -b_0 e^s \frac{\partial E_s}{\partial y} + D_0(s)e^{2s}\frac{\partial^2 E_s}{\partial y^2} - 1 - D_0(s)e^{2s}\left(\frac{\partial E_s}{\partial y}\right)^2 \qquad (39)$$

In this form, the coefficients in the equation are independent of the spatial variable y. In particular, this ensures that the solutions corresponding to the periodic initial conditions will be periodic with the same period (the initial conditions are the conditions at the zero scale, $s = 0$). As follows from Eq. (38), at the initial scale y is equal to x, so that rescaling (38) does not disturb the initial conditions.

For the sample calculations, we will consider the initial diffusion coefficients $d_{s=0}(x)$ defined for $0 \leq x \leq 1$ and then continued periodically on the infinite axis. Numerically, this is achieved by "gluing" the points $x = 0$ and $x = 1$, so that no boundary conditions are required. Upscaling diffusion in a periodic medium is one of the common problems in both volume averaging and the homogenization theory. The two kinds of initial conditions considered here are sinusoidal,

$$d_0(x) = 1 + \varepsilon + \sin 2\pi x, 0 \leq x \leq 1, \qquad (40)$$

and stepwise,

$$d_0(x) = \varepsilon + H(x - 0.25) - H(x - 0.75), 0 \leq x \leq 1 \qquad (41)$$

Here H is the Heaviside stepwise function. Condition (41) is a simplified version of conditions (24) or (25), with equal volumes of the high-permeable and low-permeable media (either channels and blocks or cavities and borders). In simulations, coefficient b_0 is taken to be zero, and D_0 is always constant.

The numerical solution of Eq. (39) was produced in Matlab by the method of lines (Schiesser, 1991). The equation was discretized into a system of ordinary differential equations for variables $E^k(s) = E_s(y_k)$, with $y_k = (k - 1)y_{\max}/K$ (where K is the total number of discretization points). A common differential equation of the system has a form of

$$\frac{dE^k}{ds} = -b_0 e^s \frac{E^{k+1} - E^{k-1}}{\Delta y} - 1 + D_0(s)e^{2s}\frac{E^{k+1} - 2E^k + E^{k-1}}{\Delta y^2}$$
$$- D_0(s)e^{2s}\left(\frac{E^{k+1} - E^{k-1}}{\Delta y}\right)^2$$

Periodicity is ensured by setting $E^{K+1} = E^1$ and $E^0 = E^K$.

The computations on an ordinary PC were rather fast so that the number of the discretization points was not an issue. In the simulations, we took K equal to 500. Such a large number was not necessary for the convergence of the numerical solution of the system of differential equations, but, rather, because of the following problem. After the solution is obtained, it has to be rescaled back to the original variable x, according to Eq. (38). Since the rescaled values $x_k = e^{-s}y_k$ do not have to coincide with the original grid $k'\Delta x$, the linear interpolation was applied to compute the dependence in these points. This interpolation is preferential over the spline interpolation since it does not create artifacts around the zones of high gradients. For the accuracy of the linear interpolation, a large number of the discretization points was required.

3.4.2. Results of calculations

The results of sample calculations are shown in Figs. 2 and 3 for the initial conditions (40) and (41), correspondingly. We plot the dependence of the value $G_s = e^s d_s$ on x and s. The multiplier e^s is introduced to make the resulting expression tending to a constant as s increases. This tendency follows from the dimension of the diffusion coefficient, cf. the Eq. (34) and the discussion above it. According to this dimensional analysis, if x and t are scaled by
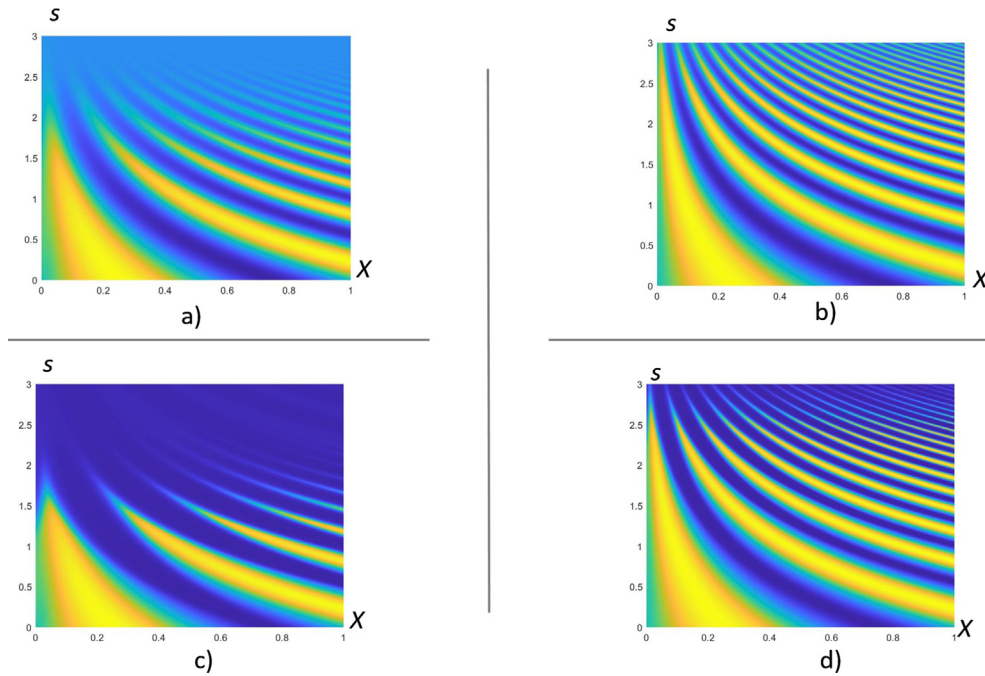
Fig. 2. Solution of the upscaling equation for the diffusion coefficient, for the initial condition (40): a) $\varepsilon = 0.5$, $D_0 = 10^{-3}$; b) $\varepsilon = 0.5$, $D_0 = 10^{-4}$; c) $\varepsilon = 10^{-4}$, $D_0 = 10^{-3}$; d) $\varepsilon = 10^{-4}$, $D_0 = 10^{-4}$.

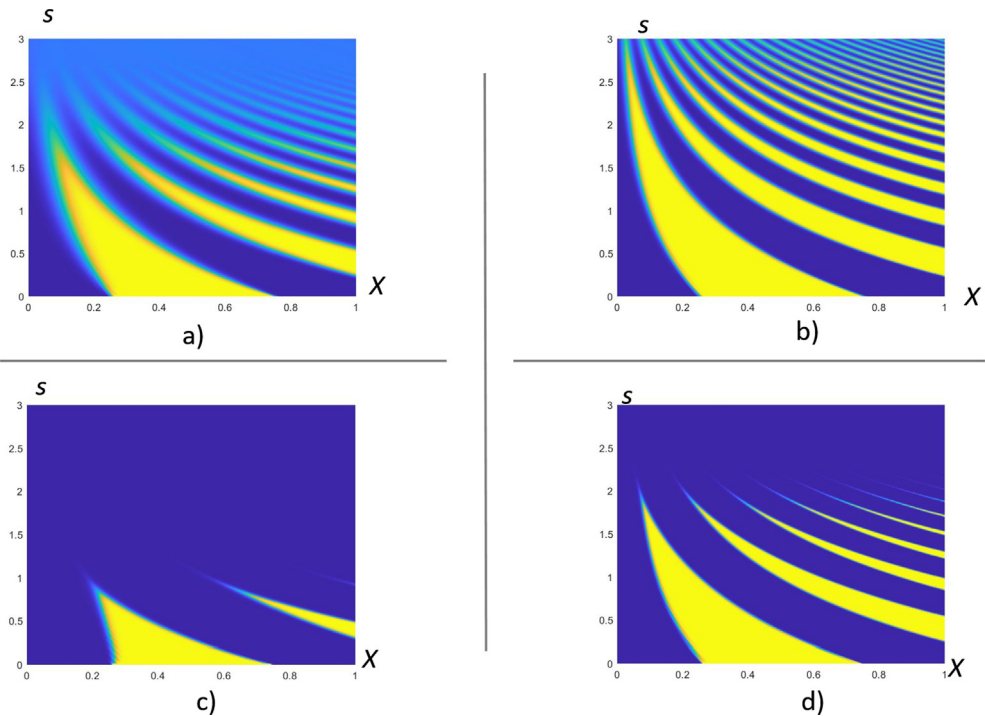**Fig. 3.** Solution of the upscaling equation for the diffusion coefficient, for the initial condition (41): a) $\varepsilon = 0.5$, $D_0 = 10^{-3}$; b) $\varepsilon = 0.5$, $D_0 = 10^{-4}$; c) $\varepsilon = 10^{-4}$, $D_0 = 10^{-3}$; d) $\varepsilon = 10^{-4}$, $D_0 = 10^{-4}$.

e^{-s}, a constant diffusion coefficient should also be scaled by e^{-s}. If a non-constant diffusion coefficient tends to a constant under scaling, this constant should obey the same rule.

In Figs. 2 and 3, the values of G_s are shown by color. The largest values correspond to the yellow, and the smallest to the deep blue color. The value of x varies on the plots over the period of the initial condition: $0 \leq x \leq 1$. The value of s changes from 0 to 3, which corresponds to ca. 20-fold zoom-out.

The computations are carried out for the two values of the smoothening (averaging) coefficient D_0 : smaller smoothening (10^{-4}) and larger smoothening (10^{-3}). The two values of ε were considered in the initial conditions (40), (41). The value of $\varepsilon = 10^{-4}$ corresponds to the case where the diffusion coefficient becomes almost zero at certain intervals (as for example in the porous medium), while $\varepsilon = 0.5$ corresponds to the case where

the diffusion coefficient varies within a single order of magnitude. Each Fig. 2,3 contains four plots corresponding to the two different values of D_0 and the two different values of ε.

A common feature of all eight plots is the behavior of the zones with high values of G_s. As s increases, these zones tend to $x = 0$, due to scaling. They also get narrower. This cannot be explained by smoothing alone since the zones diminish and eventually disappear even for very small coefficients D_0. The effect of narrowing is, mainly, produced by the term $-2D_0(s)\frac{1}{d_s(x)}\left(\frac{\partial d_s}{\partial x}\right)^2$ in Eq. (19). This term results in a rapid decrease of d_s where this coefficient is small and the gradients are sharp. This is confirmed by the fact that the convergence to a constant is faster for the initial condition (41) containing an abrupt variation of the d_s.

The asymptotic value of G_s at high s is close to the smallest initial value. It is equal to 0.72 for $\varepsilon = 0.5$ and $2 \cdot 10^{-4}$ for $\varepsilon = 10^{-4}$. This corresponds to the physical intuition, according to which low-permeable zones "kill" the diffusion in one dimension. In three dimensions, it is possible to avoid obstacles by going around them. However, this possibility is excluded in the factorized solutions for the concentration, like (26). This indicates, again, that upscaling of the diffusion coefficient depends not only on the medium but also on a selected solution for the flow. That is why the asymptotic values of the diffusion coefficient are, apparently, lower than those produced by the empirical formula developed previously, as well as the numerical simulations based on particle Brownian motion (Alvarez-Ramirez et al., 2016).

The obtained asymptotical value of the diffusion coefficient is worth comparing with the value obtained by application of the VAT. We refer to the book of Whitaker (1999); more precisely, to Eqs. (1.4.58a-c), (1.4.62), (1.4.64), and problem 1.20 from this book. We compare the asymptotic value D_{eff} from the VAT with our solution corresponding to the initial condition (41), with the infinitesimal value of ε. Our approach, in agreement with physical intuition, provides the asymptotical value of the diffusion coefficient to be equal to zero. In the framework of the VAT, under constant porosity and absence of the chemical reactions, the effective diffusion coefficient in VAT is given by

$$D_{eff} = D\left(\boldsymbol{I} + \frac{1}{V_\gamma}\int_{A_{\gamma k}}\boldsymbol{n}_{\gamma k}\boldsymbol{b}dA\right)$$

Here D is the original diffusion coefficient in the openings; V_γ is the averaging volume of the flowing (gamma) phase; $\boldsymbol{n}_{\gamma k}$ is the normal vector at the interface; and $\boldsymbol{b}$ is the vector obeying the following conditions:

$$\nabla^2\boldsymbol{b} = 0$$

$$-\boldsymbol{n}_{\gamma k}\cdot\nabla\boldsymbol{b} = \boldsymbol{n}_{\gamma k}$$

The last condition is stated on the surface $A_{\gamma k}$ between the flowing (gamma) and the solid (k) phases.

It is easy to check that, for 1D problems, $\boldsymbol{b} = -x(\text{mod}1)$, and D_{eff} is zero (the same result may more easily be obtained from Eq. (4) of problem 1.23, Whitaker (1999)). Thus, in this case both VAT and our approach agree.

3.4.3. Discussion

The similarity of the asymptotic diffusion coefficients between this work and VAT, demonstrated at the end of the previous subsection, is a part of a more general question about the similarities and distinctions between the theory developed in this work and the previously developed homogenization theories. This question is briefly discussed in this section. For simplicity of discussion, we refer to the VAT, although many statements are relevant to other averaging schemes.

As for now, VAT is much more developed and considers many more cases than the current approach, thanks to its over 60-years history. It is too early to compare the power and capacities of both approaches. Many problems, considered and resolved in the framework of VAT (e.g. unsteady-state diffusion with nonlinear chemical reactions, and much more) still need to be studied in the framework of the continuous upscaling.

An obvious advantage of the method developed in this work and our previous paper (Shapiro, 2021) is that it does not only provide the relationships between the "least" scales but shows the continuous smoothing transformation through the whole axis of scales. This raises a question, whether the final result of this transformation, the value of diffusion coefficient at $s \to \infty$, is equal to, or, at least, somehow corresponds to the value of diffusion coefficient obtained with the VAT or other homogenization theories. This question is nontrivial, since, unlike VAT, in the continuous upscaling approach the values of the effective diffusion coefficient depend on the solution for concentration on the fine scale. The upscaling equations (35) and (37) are different for the solutions (27) and (28), and cannot be reduced to each other. Moreover, for some solutions, which cannot be factorized as described in Section 3.3.2, a separate equation for the diffusion coefficient may not exist (or, at least, has not been derived). Though, the preliminary considerations described at the end of the previous subsection indicate that the final upscaled diffusion coefficients may be the same. A more detailed comparison of the asymptotically averaged diffusion coefficient requires a separate study.

In the VAT, the independence of the effective diffusion coefficient on the fine-scale solution for the concentration is obtained by careful selection of the assumptions about the flow on the micro-level, and an elaborate procedure for the scale separation. Such set of assumptions is yet to be developed for the continuous upscaling. To the best of our knowledge, the same question has not been studied, either for comparison of the VAT and the weighted averaging. At least, some studies (e.g. Baveye, 2004) indicate that the result of such averaging may depend on the chosen weight function. Some methods and problems of averaging, with multiple-scale heterogeneities, may result in a necessity to solve coupled systems of equations in different scales (Panfilov and Rasoulzadeh, 2013). This may be interpreted as a form of the dependence of the transport coefficients on the course scale on the solution of the fine scale. This confirms, at least, that separation of the scales is a necessary condition for the independence of the coarse-scale transport coefficients of the particular solution for concentration on the fine scale.

4. Conclusions

In this paper, we apply the theory of continuous upscaling of transport equations developed by Shapiro, 2021, to upscaling the stationary diffusion equation in a heterogeneous medium. As in the previous paper, we introduce the continuous line of scales counted by the parameter s, minus the logarithm of the scaling coefficient. The value of s increases from a finer to a coarser scale. Partial differential equations are derived to upscale densities (of mass, momentum, energy...), fluxes, and other parameters of the transport of matter.

Application of the theory requires its generalization onto three dimensions. The details of such generalization may be nontrivial. While the generalization of the upscaling equations for densities is straightforward, the equation for fluxes requires some additional consideration. We have derived a condition for upscaling the fluxes, which guarantees the preservation of the form of a general transport equation. This condition is shown to be compatible with upscaling of the expression $\rho\boldsymbol{v}$. This expression plays a double role,

as a density of momentum and as a flux in the mass conservation law. It is shown that the condition for upscaling fluxes does not contradict to upscaling of $\rho\boldsymbol{v}$ as a density. An algebra of upscaling operators has been developed and applied to derive the upscaling equations for velocity and non-convective fluxes.

The developed theory has been applied to upscaling of the diffusion equation in a heterogeneous medium. In one dimension, an exact equation for upscaling the diffusion coefficient is derived. This equation is only valid for the 1D diffusion equation without a source term, while the presence of such a term results in a different upscaling law.

The 1D upscaling equation for the diffusion coefficient is solved and studied numerically for periodic initial conditions. It is demonstrated that the expression $e^s d_s$ tends to a constant as s increases. This constant is close to a minimum value among the diffusion coefficients in the initial distribution. Hence, the presence of low-permeable zones hinders diffusion on the whole axis. Convergence under upscaling happens by smoothing the coefficient, as well as by narrowing the zones with high values of the diffusion coefficients.

In three dimensions, the picture is much more complicated. A more or less complete study is possible for the case where the diffusion coefficient may be factorized into a product of the coefficients, each depending on its own spatial coordinate. In such cases, the diffusion equation allows for factorized solutions. For a wide class of such solutions, it is demonstrated that the upscaling equation for the diffusion coefficient is similar to the 1D case. However, for other solutions, the upscaling equation may become different. For a general diffusion equation in 3D, it seems to be impossible to derive an upscaling law for the diffusion coefficient.

Further development of the theory may be governed by the fundamental problems, described above (e.g. section 3.4.3), as well as engineering and industrial needs. The last developments may involve inclusion of the unsteady flows, chemical reactions, as well as consideration of the multiscale heterogeneous processes, like turbulence or flows in natural porous media.

CRediT authorship contribution statement

Alexander A. Shapiro: Conceptualization, Formal analysis, Investigation, Methodology, Project administration, Software, Supervision, Validation, Writing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Appendix A. A form of the diffusion coefficient corresponding to the factorized concentration

The goal of this appendix is to solve the problem formulated in Section 3.3.2: What is the general form of the diffusion coefficient admitting for the factorized solutions (26) for the concentration? We start with the equation (16):

$$\frac{\partial c}{\partial x_i}\frac{\partial \ln d}{\partial x_i} + \frac{\partial^2 c}{\partial x_j^2} = 0$$

Substitution of the factorized solution and utilization of the rules of differentiation (rd) reduces it to

$$\frac{1}{c^i}\frac{\partial c^i}{\partial x_i}\frac{\partial \ln d}{\partial x_i} + \frac{1}{c_k}\frac{\partial^2 c^k}{\partial x_k^2} = 0$$

For simplification of the equation we make a substitution (with a summation by i):

$$\ln d = \ln d^i(x_i) + \ln d^0(\boldsymbol{x})$$

Here coefficients $\ln d^l(x_I)$ are solutions of the corresponding individual equations:

$$\frac{\partial c^l}{\partial x_I}\frac{\partial \ln d^l}{\partial x_I} + \frac{\partial^2 c^l}{\partial x_I^2} = 0$$

The corresponding terms are canceled out, and the rest coefficient $\ln d^0$ obeys the homogeneous equation

$$\frac{1}{c^i}\frac{\partial c^i}{\partial x_i}\frac{\partial \ln d^0}{\partial x_i} = 0$$

Introduce the new set of independent variables

$$X_I = \int \frac{c^i dx_i}{\frac{\partial c^i}{\partial x_i}}$$

Then the last equation becomes

$$\frac{\partial \ln d^0}{\partial X_i} = 0$$

Its particular solution is, for arbitrary constants γ_i,

$$\ln d^0 = \ln d^0(\gamma^i X_i), \gamma^1 + \gamma^2 + \gamma^3 = 0 \tag{42}$$

A general solution d_{gen} is a closure of the space of arbitrary linear combinations of the form (42) with the different sets of γ^i.

Finally, the diffusion coefficient is

$$d = \ln d_{gen}d^1(x_1)d^2(x_2)d^3(x_3)$$

Thus, a factorized solution for the concentration is possible the diffusion coefficient is a product of the terms $d^l(x_I)$ and an additional term d_{gen}, which "spoils" its factorization.

Appendix B. . Derivation of the equations for the components of the factorized diffusion coefficient. Case 1

In this appendix, we derive the individual upscaling equations for the components $d^i(x_i)$ of the factorized diffusion coefficient and concentration. The starting points for the derivation are the general diffusion upscaling equation (22); Eqs. (23) and (26) for factorization of the diffusion coefficients and the concentration; and Eq. (27) for the individual components of the concentration $c^i(x_i)$.

In the following computations, we will use the simple rules for differentiation of the factorized quantities:

$$\frac{\partial c_s}{\partial x_I} = \frac{c_s}{c_s^I}\frac{\partial c_s^I}{\partial x_I}; \frac{\partial^2 c_s}{\partial x_I^2} = \frac{c_s}{c_s^I}\frac{\partial^2 c_s^I}{\partial x_I^2}; \frac{\partial^2 c_s}{\partial x_I \partial x_K} = \frac{c_s}{c_s^I c_s^K}\frac{\partial c_s^I}{\partial x_I}\frac{\partial c_s^K}{\partial x_K}(I \neq K); \tag{43}$$

Similarly,

$$\frac{\partial d_s}{\partial x_I} = \frac{d_s}{d_s^I}\frac{\partial d_s^I}{\partial x_I}; \left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s = \frac{d_s}{d_s^I}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i \tag{44}$$

Here $\boldsymbol{u}_s^I$ is a 1D version of the upscaling operation $\boldsymbol{u}_s$ as defined by Eq. (31).

The derivation starts with equation (22) represented in the form of

$$\frac{\partial Z^k}{\partial x_k} = 0; Z^K = \frac{\partial c_s}{\partial x_K}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s\right)d_s - 2D_0(s)\frac{\partial d_s}{\partial x_i}\frac{\partial^2 c_s}{\partial x_i \partial x_K} \quad (K = 1, \cdots, 3) \quad (45)$$

First, we transform the expression for Z^K, substituting the factorizing equations (26) and (23):

$$Z^K = \frac{c_s}{c_s^K}\frac{d_s}{d_s^i}\frac{\partial c_s^K}{\partial x_K}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i - 2D_0(s)\sum_{i\neq K}\frac{d_s}{d_s^i}\frac{\partial d_s^i}{\partial x_i}\frac{c_s}{c_s^i c_s^K}\frac{\partial c_s^i}{\partial x_i}\frac{\partial c_s^K}{\partial x_K}$$

$$- 2D_0(s)\frac{d_s}{d_s^K}\frac{\partial d_s^K}{\partial x_K}\frac{c_s}{c_s^K}\frac{\partial^2 c_s^K}{\partial x_K^2}$$

The second derivative in the last addendum may be expressed from the individual diffusion equation (27), resulting in

$$Z^K = d_s\frac{c_s}{c_s^K}$$

$$\times \frac{\partial c_s^K}{\partial x_K}\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i - 2D_0(s)\sum_{i\neq K}\frac{1}{d_s^i}\frac{\partial d_s^i}{\partial x_i}\frac{1}{c_s^i}\frac{\partial c_s^i}{\partial x_i} + 2D_0(s)\frac{1}{\left(d_s^K\right)^2}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2\right]$$

We substitute this expression to Eq. (45):

$$\frac{c_s}{c_s^k}\frac{d_s}{d_s^k}\frac{\partial}{\partial x_k}\left\{\left(d_s^k\frac{\partial c_s^k}{\partial x_k}\right)\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i - 2D_0(s)\sum_{i\neq k}\frac{1}{d_s^i}\frac{\partial d_s^i}{\partial x_i}\frac{1}{c_s^i}\frac{\partial c_s^i}{\partial x_i} + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_k}\right)^2\right]\right\}$$

$$= 0$$

The multipliers $\frac{c_s}{c_s^k}\frac{d_s}{d_s^k}$ are exchanged with the derivative $\frac{\partial}{\partial x_k}$, since they are independent of x_k. If this derivative is applied to the term $d_s^k\frac{\partial c_s^k}{\partial x_k}$ (the term in round parentheses in the last equation), the result is zero, due to Eq. (27). Applying the formula for derivative of the product (of the terms in the round and square parentheses), we obtain:

$$\frac{c_s}{c_s^k}d_s\frac{\partial c_s^k}{\partial x_k}$$

$$\times \frac{\partial}{\partial x_k}\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i - 2D_0(s)\sum_{i\neq k}\frac{1}{d_s^i}\frac{\partial d_s^i}{\partial x_i}\frac{1}{c_s^i}\frac{\partial c_s^i}{\partial x_i} + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_K}\right)^2\right]$$

$$= 0$$

This equation may be greatly simplified if we notice that the terms like $\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i$ and $\frac{1}{d_s^i}\frac{\partial d_s^i}{\partial x_i}\frac{1}{c_s^i}\frac{\partial c_s^i}{\partial x_i}$ are independent of x_k, if $i\neq k$. These terms are reduced by the derivative $\frac{\partial}{\partial x_k}$. Excluding them and reducing the common multiplier $c_s d_s$, we obtain:

$$\frac{1}{c_s^k}\frac{\partial c_s^k}{\partial x_k}\frac{\partial}{\partial x_k}\left[\frac{1}{d_s^k}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^k\right)d_s^k + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_K}\right)^2\right] = 0$$

The different addenda in the last equation depend on a single variable x_k. It is obeyed if they are equal to zero individually, or if

$$\frac{\partial}{\partial x_K}\left\{\frac{1}{d_s^k}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^k + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_K}\right)^2\right\} = 0 (K = 1, \cdots 3)$$

Vice versa, if the last equation is valid, then back substitution results in the validity of Eq. (45) equivalent to the 3D upscaling equation (22) for the factorized case.

Appendix C. . Derivation of the equations for the components of the factorized diffusion coefficient. Case 3

The derivations of this appendix are similar to Appendix B, with some modifications. Again, Eq. (22) forms the basis for the derivation. The rules (43) are substituted by

$$\frac{\partial c_s}{\partial x_l} = \frac{\partial c_s^l}{\partial x_l}; \frac{\partial^2 c_s}{\partial x_l^2} = \frac{\partial^2 c_s^l}{\partial x_l^2}; \frac{\partial^2 c_s}{\partial x_l \partial x_K} = 0 (I\neq K); \quad (46)$$

The rules (44) remain unchanged.

Introducing Z^K like in Eq. (45) and applying the rules (46), (44), we transform them into the form

$$Z^K = \frac{d_s}{d_s^i}\frac{\partial c_s^K}{\partial x_K}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i - 2D_0(s)\frac{d_s}{d_s^K}\frac{\partial d_s^K}{\partial x_K}\frac{\partial^2 c_s^K}{\partial x_K^2}$$

The second derivative in the last addendum of this equation may be expressed from Eq. (30) represented in the form of

$$\frac{\partial^2 c_s^i}{\partial x_l^2} = H^I - \frac{1}{d_s^I}\frac{\partial d_s^I}{\partial x_l}\frac{\partial c_s^I}{\partial x_l}:$$

$$Z^K = d_s\frac{\partial c_s^K}{\partial x_K}\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i + 2D_0(s)\frac{1}{\left(d_s^K\right)^2}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2\right] - 2D_0(s)H^K$$

$$\times \frac{d_s}{d_s^K}\frac{\partial d_s^K}{\partial x_K}$$

Substitution back to Eq. (45) and the transformation similar to the previous appendix results in:

$$\frac{d_s}{d_s^k}\frac{\partial}{\partial x_k}\left\{\left(d_s^k\frac{\partial c_s^k}{\partial x_k}\right)\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_k}\right)^2\right] - 2D_0(s)H^k\frac{\partial d_s^k}{\partial x_k}\right\}$$

$$= 0$$

When the derivative $\frac{\partial}{\partial x_k}$ is applied to the first term in the braces, it should be accounted for the fact that $\frac{\partial}{\partial x_k}\left(d_s^k\frac{\partial c_s^k}{\partial x_k}\right)$ is equal to H^k, due to Eq. (27). The result looks rather ugly:

$$d_s H^k\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_k}\right)^2\right] + d_s\frac{\partial c_s^k}{\partial x_k}$$

$$\times \frac{\partial}{\partial x_k}\left[\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_K}\right)^2\right] - \frac{d_s}{d_s^k}$$

$$\times \frac{\partial}{\partial x_k}\left[2D_0(s)H^k\frac{\partial d_s^k}{\partial x_k}\right]$$

$$= 0$$

However, it may be greatly simplified with the help of the following two observations. First, it may be noticed that in the first addendum the following expression cancels out, since the sum of H^k is equal to zero:

$$H^k\frac{1}{d_s^i}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^i\right)d_s^i = 0$$

In the second addendum, the derivative $\frac{\partial}{\partial x_k}$ eliminates all the terms indexed by i, for $i\neq k$. The result, after division by $\frac{\partial c_s^k}{\partial x_k}$, assumes a more bearable form

$$\frac{\partial}{\partial x_k}\left[\frac{1}{d_s^k}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^k\right)d_s^k + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_K}\right)^2\right] + 2D_0(s)H^k$$

$$\times \frac{1}{d_s^k\frac{\partial c_s^k}{\partial x_k}}\left[\frac{1}{d_s^k}\left(\frac{\partial d_s^k}{\partial x_k}\right)^2 - \frac{\partial^2 d_s^k}{\partial x_k^2}\right]$$

$$= 0$$

Finally, to exclude concentration from the last addendum, we integrate Eq. (30):

$$d_s^K \frac{\partial c_s^K}{\partial x_K} = H^K \int d_s^K dx_K,$$

which gives:

$$\frac{\partial}{\partial x_k}\left[\frac{1}{d_s^k}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^k\right)d_s^k + 2D_0(s)\frac{1}{\left(d_s^k\right)^2}\left(\frac{\partial d_s^k}{\partial x_k}\right)^2\right] + 2D_0(s)\frac{1}{\int d_s^k dx_k}\left[\frac{1}{d_s^k}\left(\frac{\partial d_s^k}{\partial x_k}\right)^2 - \frac{\partial^2 d_s^k}{\partial x_k^2}\right] = 0$$

For each k, the corresponding terms depend only on x_k their sum is equal to zero if they are equal to zero individually:

$$\frac{\partial}{\partial x_k}\left[\frac{1}{d_s^K}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^K\right)d_s^K + 2D_0(s)\frac{1}{\left(d_s^K\right)^2}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2\right] + 2D_0(s)\frac{1}{\int d_s^K dx_K}\left[\frac{1}{d_s^K}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2 - \frac{\partial^2 d_s^K}{\partial x_K^2}\right] = 0\#$$

(47)

This is an equation for individual diffusion coefficients d_s^K independent of the concentration. What is more wonderful, it is also independent of constants H^k. If the integration in the last term is seen as an obstacle, the equation may be expressed in the equivalent (but still cumbersome) form

$$d_s^K = \frac{\partial}{\partial x_K}\left\{\frac{\left(2D_0(s)\left[\frac{1}{d_s^K}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2 - \frac{\partial^2 d_s^K}{\partial x_K^2}\right]\right)}{\frac{\partial}{\partial x_K}\left[\frac{1}{d_s^K}\left(\frac{\partial}{\partial s} - \boldsymbol{u}_s^K\right)d_s^K + 2D_0(s)\frac{1}{\left(d_s^K\right)^2}\left(\frac{\partial d_s^K}{\partial x_K}\right)^2\right]}\right\}$$

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