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Macrodispersivity for transport in arbitrary nonuniform flow fields: Asymptotic and preasymptotic results

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[1] We use homogenization theory to investigate the asymptotic macrodispersion in arbitrary nonuniform velocity fields, which show small-scale fluctuations. In the first part of the paper, a multiple-scale expansion analysis is performed to study transport phenomena in the asymptotic limit $\varepsilon \ll 1$, where ε represents the ratio between typical lengths of the small and large scale. In this limit the effects of small-scale velocity fluctuations on the transport behavior are described by a macrodispersive term, and our analysis provides an additional local equation that allows calculating the macrodispersive tensor. For Darcian flow fields we show that the macrodispersivity is a fourth-rank tensor. If dispersion/diffusion can be neglected, it depends only on the direction of the mean flow with respect to the principal axes of anisotropy of the medium. Hence the macrodispersivity represents a medium property. In the second part of the paper, we heuristically extend the theory to finite ε effects. Our results differ from those obtained in the common probabilistic approach employing ensemble averages. This demonstrates that standard ensemble averaging does not consistently account for finite scale effects: it tends to overestimate the dispersion coefficient in the single realization. *INDEX TERMS:* 1832 Hydrology: Groundwater transport; 1869 Hydrology: Stochastic processes; 5139 Physical Properties of Rocks: Transport properties; *KEYWORDS:* homogenization theory, two-scale analysis, nonuniform flow, upscaling, macrodispersivity

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1. Introduction

[2] The conductivity of natural porous media can vary over several orders of magnitude within the same formation. Flow through such media might be highly heterogeneous with characteristic lengths varying from microscopic up to the macroscopic scale and it is impossible to incorporate explicitly any detailed structure in a macroscopic analysis. On the other hand, the small-scale heterogeneity may have an important influence on the system behavior. A possible way to overcome this problem is to describe explicitly only the large-scale heterogeneities, while the small-scale variability is described implicitly in terms of its effects on large-scale quantities. The problem arises, how to compute these large-scale parameters.

[3] For solute transport in a velocity field that is heterogeneous on the small scale, this problem reduces to the identification of a macrodispersive term. For Darcian flow through porous media this problem has been investigated in the case of large-scale uniform flow fields by several authors and with different techniques [see, e.g., *Gelhar and Axness*, 1983; *Dagan*, 1984; *Kitanidis*, 1988]. Only recently work has been published, which investigates the large-scale behavior of a solute in a nonuniform velocity field [*Indelman and Dagan*, 1999; *Dagan and Indelman*, 1999; *Attinger et al.*, 2001]. However, the results are limited to relatively simple nonuniform flow configurations, such as radial flow or dipole flow. In a slightly different context, *Neuman* [1993], *Guadagnini and Neuman* [2001] also consider transport in a nonuniform flow studying the possibility of incorporating nonlocal effects in the transport parameters by conditional moments, whereas *Dagan et al.* [1996] deal with a flow, which is not uniform in time.

[4] We consider a velocity field that shows two distinct scales of variation: a mesoscopic scale, ℓ, at which the medium is heterogeneous, and a macroscopic scale, L, at which the transport phenomena are observed. At first we focus our attention on the case of well-separated scale: the small-scale heterogeneities have a typical length much smaller than the observation scale, $\varepsilon := \ell/L \to 0$. Homogenization theory provides a very natural and elegant way to solve this problem; it allows us to develop a rigorous analysis based on spatially averaged quantities. If the scale separation does not hold, the upscaling problem is not well posed in the sense that the macroscopic mean concentration may not obey an advection-dispersion equation [see, e.g., *Smith and Schwartz*, 1980]. In this case the behavior of the system is normally described by means of an advective-dispersion equation in a probabilistic sense, thus describing the behavior of the ensemble averaged concentration. Due to the lack of ergodicity, the ensemble-averaged solution is not suitable to predict the transport in a single realization. To describe these phenomena we can simply extend our formalism to describe the case of small but finite ε. The price to pay is the loss of rigor, the derivation being heuristic and needing some ad hoc, though well motivated, assumptions.

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[5] Homogenization theory has been widely applied to purely diffusive problems [e.g., flow in porous media), where the elliptic nature of the operator makes the analysis relatively easy [see, e.g., *Papanicolaou and Varadhan*, 1981; *Sanchéz-Palencia*, 1980]. Extension to transport problems with a zero-mean advective term have been also studied in the context of turbulent flow [see, e.g., *Avellaneda and Majda*, 1990]. Homogenization of transport with a nonzero mean drift is a more difficult task because the hyperbolic advective term complicates the classical purely elliptical problem. Indeed, the advective and dispersive parts may exhibit different characteristic lengths and/or times. Few authors treated this problem and we are not aware of any application to determine the macrodispersion tensor. Previous works apply homogenization to the study of Taylor dispersion [*Rubinstein and Mauri*, 1986; *Mei*, 1992; *Auriault and Adler*, 1995]. Homogenization theory basically employs a two-scale asymptotic expansion in a small parameter ε, typically the ratio between the characteristic mesoscopic and macroscopic lengths. Since we are interested in the heterogeneity-enhanced dispersion, we rescale the time dispersively. Thus our analysis is a straightforward consequence of the kinematic relation among position, time and velocity; in this our derivation of the macroscopic equation differs qualitatively from the previous works. The concentration is expanded in a power series in terms of ε, i.e., $C = C_0 + \varepsilon C_1 + \varepsilon^2 C_2 + \ldots$, which represents a formal point of the analysis in the sense that a priori there is no reason that it converges and that one is allowed to truncate it neglecting higher-order terms. Apart from postulating this expansion (ansatz), our approach is rigorous.

[6] The paper proceeds as follows. In section 2 we state the problem and give the definition of a two-scale function. In section 3 we apply a multiple-scale analysis to the diffusive-advective equation in order to identify the macrodispersion coefficients. In section 4 we restrict our attention to porous media; explicit results obtained by lowest-order perturbation theory are compared with previous works. In section 5, we extend our approach to the case of finite ε. Explicit results are presented. Finally, in section 6 we summarize the main findings of the paper.

2. Statement of the Problem

[7] In this paper we investigate the macroscopic behavior of a conservative solute transported in a heterogeneous velocity field. The solute concentration obeys a mesoscopic advection-dispersion equation of the form

$$\frac{\partial}{\partial \tau} C(\boldsymbol{\xi}, \tau) + \boldsymbol{u}(\boldsymbol{\xi}) \cdot \nabla_{\xi} C(\boldsymbol{\xi}, \tau) \nabla_{\xi} \cdot \left[\mathbf{D} \nabla_{\xi} C(\boldsymbol{\xi}, \tau) \right] = 0, \qquad (1)$$

where the velocity field $\boldsymbol{u}$ depends on the spatial variable $\boldsymbol{\xi}$. The presence even of a small dispersion cannot be neglected at all, since it changes qualitatively the nature of the solution (spreading of the plume). In most applications in heterogeneous porous media, one is interested in the advection-dominated behavior. For the sake of simplicity, we assume that the small-scale dispersion tensor D_{ij} is constant in space and isotropic, i.e., $D_{ij} = D\delta_{ij}$. However, extension of our analysis to a more realistic velocity dependent dispersivity is straightforward. The velocity field $\boldsymbol{u}(\boldsymbol{\xi})$ exhibits two typical length scales of variation: a scale ℓ at which the heterogeneous properties of the medium fluctuate and the transport length scale L at which the transport behavior is observed. If these two length scales are well separated, i.e., in the limit $\ll L$, it is convenient to consider the velocity field as a two-scale function and investigate the transport problem with a two-scale expansion technique.

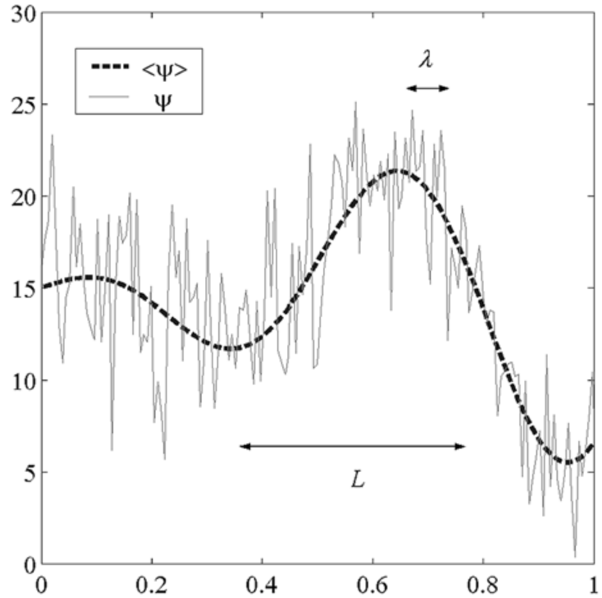

Figure 1. Two-scale function ψ and its mean value $\langle \psi \rangle = \psi^*$.

2.1. Two-Scale Functions

[8] In general, a two-scale function can be written as $\psi(\boldsymbol{\xi}) = \psi(s(\boldsymbol{\xi}), r(\boldsymbol{\xi}))$, where $r(\boldsymbol{\xi})$ is the "rapidly" varying part of ψ representing the mesoscopic fluctuations and $s(\boldsymbol{\xi})$ the slowly varying part describing the macroscopic trend. We introduce at each point of the domain an appropriate averaging volume V, on which we define an average operator

$$\langle \ \rangle := \frac{1}{|\mathrm{V}|} \int_{\mathrm{V}} (\ ) d\boldsymbol{\xi}, \qquad (2)$$

$|\mathrm{V}| := \int_{\mathrm{V}} d\boldsymbol{\xi}$ is the volume of V. Therefore ψ can be split into a smoothly varying mean value $\psi^* := \langle \psi \rangle$ and a zero-mean residual $\tilde{\psi} := \psi - \psi^*$. A schematic representation of a two-scale function is given in Figure 1 for the one-dimensional case. Notice that $\psi(\boldsymbol{\xi}) = \psi^*(s(\boldsymbol{\xi})) + \tilde{\psi}(r(\boldsymbol{\xi}))$ represents a special case, in which the residual exhibits small-scale fluctuations only. This representation is inadequate in many situations, e.g., for the description of the velocity field in a nonuniform Darcian flow where the small-scale fluctuations depend also on the nonuniform mean velocity.

[9] We introduce two different spatial variables: $\boldsymbol{y}$ for small-scale variations, and $\boldsymbol{x}$ for large-scale variations. The smoothly varying part depends on $\boldsymbol{x}$, $s = s(\boldsymbol{x} := \boldsymbol{\xi})$, and the rapidly varying one on $\boldsymbol{y}$, $r = r(\boldsymbol{y} := \boldsymbol{\xi})$, and the two-scale function can be represented as a function of the two new variables: $\psi(\boldsymbol{\xi}) = \psi(\boldsymbol{x}, \boldsymbol{y})$. Note that at any point $\boldsymbol{\xi} \equiv \boldsymbol{x} \equiv \boldsymbol{y}$. $\boldsymbol{x}$ and $\boldsymbol{y}$ are simply labels applied to the physical space variable $\boldsymbol{\xi}$ to indicate that one component of ψ varies rapidly with $\boldsymbol{\xi}$, whereas the other varies slowly. These spatial variables can be naturally made dimensionless by scaling $\boldsymbol{x} = L \cdot \hat{\boldsymbol{x}}$ and $y = \ell \cdot \hat{\boldsymbol{y}}$, where L is the characteristic

macroscopic length, and ℓ the characteristic mesoscopic one. Since $\boldsymbol{x} \equiv \boldsymbol{y}$ the two dimensionless spatial variables are related by $\hat{\boldsymbol{y}} = \hat{\boldsymbol{x}}/\varepsilon$, where we defined the dimensionless number $\varepsilon := \ell/L$.

[10] Any distance can be measured in units of the small length ℓ (by the variable $\hat{\boldsymbol{y}}$) or equivalently in units of the large length L (by the variable $\hat{\boldsymbol{x}}$). The infinitesimal increments of the dimensionless variables are related by $d\hat{\boldsymbol{y}} = d\hat{\boldsymbol{x}}/\varepsilon$. If the two scales are well separated, $\varepsilon \ll 1$, an infinite increment of the mesoscopic variable corresponds to a finite increment of the macroscopic variable, whereas a finite increment of the mesoscopic variable corresponds to an infinitesimal increment of the macroscopic variable. This means that any point of the domain that is at a finite macroscopic distance from the boundary, can be regarded as infinitely far from the boundary if the distance is measured in mesoscopic lengths. Thus a problem formulated in the variable $\hat{\boldsymbol{y}}$ can be regarded as unbounded in the limit $\varepsilon \ll 1$. On the other hand, if we consider the spatial average introduced in equation (2), $\hat{\boldsymbol{x}}$ can be considered constant within the averaging volume:

$$\langle\psi(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}})\rangle = \frac{1}{|V|}\int_V (\psi^*(\hat{\boldsymbol{x}}) + \tilde{\psi}(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}}))d\hat{\boldsymbol{y}} = \psi^*(\hat{\boldsymbol{x}}). \quad (3)$$

The mean value of the multiple-scale function only depends on the macroscopic variable, i.e., $\psi^* = \psi^*(\boldsymbol{x})$.

2.2. Dimensionless Transport Equation and the Timescales

[11] The first step of a two-scale analysis is to write the transport equation in dimensionless form. The choice of the typical values to be used depends on the problem one wants to study and it is a critical point, because only a physically based scaling leads to the correct results without any additional ad hoc assumptions. Here we are interested in describing macrodispersive effects on the large-scale concentration. Therefore we adopt a macroscopic description and we scale the time diffusively, i.e., we introduce the nondimensional quantities

$$\xi = \frac{\zeta}{L}, \quad \hat{\tau} = \frac{D}{L^2}\tau, \quad \hat{\boldsymbol{u}} = \frac{\boldsymbol{u}}{U}, \quad (4)$$

where U is a typical macroscopic velocity. This is always possible provided one uses the appropriate dispersive length scale. Indeed, in studying macrodispersion the length of interest is the spread of the concentration (e.g., the size of a solute plume, the thickness of a transition front, etc.), thus one has to choose a dispersive typical length, which naturally yields a dispersive time scaling. Note that, considering the typical advective and dispersive lengths, L_A, and L_D, at a given time T, one can write $T = L_A/U = L_D^2/D$. In our opinion, this point of view provides a more physical picture than considering the advective and dispersive time related to a fixed length [*Rubinstein and Mauri*, 1986; *Mei*, 1992; *Auriault and Adler*, 1995], because it provides an immediate picture of the typical quantity.

[12] Introducing (4) into equation (1), we can write the following dimensionless equation:

$$\frac{\partial}{\partial\hat{\tau}}\hat{C}(\xi,\hat{\tau}) + \mathrm{Pe}\;\hat{\boldsymbol{u}}(\xi)\cdot\nabla_\xi\hat{C}(\xi,\hat{\tau}) - \nabla_\xi\cdot\left[\mathrm{I}\nabla_\xi\hat{C}(\xi,\hat{\tau})\right] = 0, \quad (5)$$

where $I_{ij} = \delta_{ij}$ is the identity matrix and $\mathrm{Pe} = UL/D$ the Peclet number.

[13] The velocity field is a two-scale function, thus we write $\hat{\boldsymbol{u}} = \hat{\boldsymbol{u}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$. The two spatial scales ℓ and L associated with the two spatial variables induce different timescales. Whereas the timescale described by the large-scale variable t is defined by the time needed to cover the length L, the characteristic dispersive time needed to cover the length ℓ defines a second timescale described by the small-scale variable t^ℓ. For the dimensionless temporal variables, the diffusive rescaling ($t^\ell = \ell^2/D\cdot\hat{t}^\ell$, $t = L^2/D\cdot\hat{t}$) yields $\hat{t}^\ell = \hat{t}/\varepsilon^2$ and, consequently, $d\hat{t}^\ell = d\hat{t}/\varepsilon^2$ for the infinitesimal increments. In the limit $\varepsilon \ll 1$, the latter demonstrates that an infinite increment of $\hat{t}^\ell$ corresponds to a finite increment of $\hat{t}$: the solution becomes rapidly independent of the initial conditions at mesoscopic timescales and it rapidly reaches its steady state solution with respect to mesoscopic times. Thus the large-scale transport behavior is independent of $\hat{t}^\ell$.

[14] If time is measured in dispersive units, the typical velocity depends on the observation scale: the velocity is not the same if measured at mesoscopic scale (in mesoscopic units) or at macroscopic scale (in macroscopic units). Indeed, using the kinematic relation $\boldsymbol{u} = \partial\xi/\partial\tau$ one finds

$$\boldsymbol{u} = U\cdot\frac{\partial\xi}{\partial\hat{\tau}} = U\cdot\frac{\partial\hat{\boldsymbol{x}}}{\partial\hat{t}} \equiv U\,\hat{\boldsymbol{u}}|_{\hat{x}} = \frac{U}{\varepsilon}\frac{\partial\hat{\boldsymbol{y}}}{\partial\hat{t}^\ell} \equiv \frac{U}{\varepsilon}\hat{\boldsymbol{u}}|_{\hat{y}}. \quad (6)$$

The subscript indicates whether the velocity is rescaled with respect to the macroscopic, $|_{\hat{x}}$, or to the mesoscopic, $|_{\hat{y}}$, reference quantities. For the dimensionless velocities we have

$$\hat{\boldsymbol{u}}|_{\hat{x}} = \hat{\boldsymbol{u}} = \frac{1}{\varepsilon}\hat{\boldsymbol{u}}|_{\hat{y}}. \quad (7)$$

Once the macroscopic velocity and the two spatial variable have been rescaled, one cannot arbitrarily rescale the velocity at the mesoscopic scale. If the time is rescaled dispersively, the characteristic velocity at mesoscopic scales is not U, but U/ε.

3. Two-Scale Analysis of the Transport Equation

[15] According to the relations between infinitesimal increments ($d\hat{\boldsymbol{y}} = d\hat{\boldsymbol{x}}/\varepsilon$, $d\hat{t}^\ell = d\hat{t}/\varepsilon^2$) and to the macroscopic rescaling (4) we write the gradients and the time derivatives as

$$\nabla_\xi = \nabla_{\hat{x}} + \frac{1}{\varepsilon}\nabla_{\hat{y}}. \qquad \text{and} \qquad \frac{\partial}{\partial\hat{\tau}} = \frac{\partial}{\partial\hat{t}} + \frac{1}{\varepsilon^2}\frac{\partial}{\partial\hat{t}^\ell}. \quad (8)$$

We split the velocity field in (2.11) into two parts by writing

$$\hat{\boldsymbol{u}}(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}}) = \hat{\boldsymbol{u}}^*(\hat{\boldsymbol{x}}) + \hat{\tilde{\boldsymbol{u}}}(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}}), \quad (9)$$

where $\hat{\boldsymbol{u}}^* = \langle\hat{\boldsymbol{u}}\rangle$ is the mean drift and $\hat{\tilde{\boldsymbol{u}}} = \hat{\boldsymbol{u}} - \hat{\boldsymbol{u}}^*$ the zero-mean residual defined according to the averaging operator (2). The multiple-scale function $\hat{\boldsymbol{u}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$ can be either a periodic function or a stationary random function in the mesoscopic variable $\hat{\boldsymbol{y}}$; these two cases can be handled in a analogous way in the framework of the multiple-scale analysis. In the former case the averaging volume is the period interval and ℓ the period length. If the velocity is a random function characterized by the correlation length λ, the averaging volume has to contain enough correlation lengths to satisfy $\langle \hat{\tilde{\boldsymbol{u}}} \rangle = 0$: its typical size ℓ has to be large enough compared to λ to guarantee that velocity is statistically representative, but much smaller than the macroscopic spatial scale to ensure the scale separation. Finally we notice that we do not require a stationary velocity field (statistically invariant with respect to spatial translations), but only a locally stationary one (statistically invariant with respect to small-scale spatial translations). Its average, standard deviation as well as its correlation length can be space dependent, provided they are smooth functions, i.e., functions of the variable $\hat{\boldsymbol{x}}$ only. In this sense we can assume that $\hat{\boldsymbol{u}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$ is a stationary random process in $\hat{\boldsymbol{y}}$ and a deterministic smooth function of $\hat{\boldsymbol{x}}$.

[16] By inserting (8) and (9) into the transport equation (5) we obtain

$$\partial_{\hat{t}} \hat{C} + \frac{1}{\varepsilon^2} \partial_{\hat{t}^\ell} \hat{C} + \text{Pe}\, \hat{\boldsymbol{u}}^* \cdot \nabla_{\hat{x}} \hat{C} + \text{Pe}\, \hat{\tilde{u}} \cdot \nabla_{\hat{x}} \hat{C} + \text{Pe}\, \frac{1}{\varepsilon} \hat{u} \cdot \nabla_{\hat{y}} \hat{C} - \left(\nabla_{\hat{x}}^2 \hat{C} + \frac{1}{\varepsilon} 2 \nabla_{\hat{x}} \cdot \nabla_{\hat{y}} \hat{C} + \frac{1}{\varepsilon^2} \nabla_{\hat{y}}^2 \hat{C} \right) = 0 \tag{10}$$

The third term in equation (10) represents the advection of the macroscopic concentration gradient by mean drift, thus it is obviously a macroscopic advective term and in consistency with (7) we substitute $\hat{\boldsymbol{u}}^* = \hat{\boldsymbol{u}}^*|_{\hat{x}}$. The fourth term represents the advection of the large-scale concentration gradient by the local variations of the velocity field, which acts advectively only inside an averaging volume (it has a correlation length $\lambda < \ell$); whereas the fifth term represents the advection of the small-scale concentration gradient. Both the fourth and the fifth terms are mesoscopic and according to (7), we substitute $\hat{\boldsymbol{u}} = \hat{\boldsymbol{u}}|_{\hat{y}}/\varepsilon$ and $\hat{\tilde{\boldsymbol{u}}} = \hat{\tilde{\boldsymbol{u}}}|_{\hat{y}}/\varepsilon$. Equation (10) becomes

$$\partial_{\hat{t}} \hat{C} + \frac{1}{\varepsilon^2} \partial_{\hat{t}^\ell} \hat{C} + \text{Pe}\, \hat{\boldsymbol{u}}^*|_{\hat{x}} \cdot \nabla_{\hat{x}} \hat{C} - \nabla_{\hat{x}}^2 \hat{C} + \frac{1}{\varepsilon} \left[\text{Pe}\, \hat{\tilde{\boldsymbol{u}}}\Big|_{\hat{y}} \nabla_{\hat{x}} \hat{C} - 2 \nabla_{\hat{x}} \cdot \nabla_{\hat{y}} \hat{C} \right] + \frac{1}{\varepsilon^2} \left[\text{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{C} - \nabla_{\hat{y}}^2 \hat{C} \right] = 0. \tag{11}$$

As pointed out in the previous section, we look for a solution of the form $\hat{C} = \hat{C}^{\varepsilon,\text{Pe}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t})$, that does not depend on $\hat{t}^\ell$ and we drop the time derivative with respect to mesoscopic times in (11). The next step is to expand the solution $\hat{C} = \hat{C}^{\varepsilon,\text{Pe}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t})$ around its asymptotic solution in terms of small ε

$$\hat{C}^{\varepsilon,\text{Pe}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}) = \hat{C}_0^{\text{Pe}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}) + \varepsilon \hat{C}_0^{\text{Pe}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}) + \varepsilon^2 \hat{C}_2^{\text{Pe}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}) + \ldots \tag{12}$$

where each coefficient $\hat{C}_n^{\text{Pe}}$ is now independent of ε, depending only on $\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}$, and on the Peclet number. We assume that the expansion (12) exists and converges to its asymptotic solution in the limit $\varepsilon \ll 1$. Inserting expansion (12) into (11) and collecting the terms of the same power of ε, we obtain an equation that has to be satisfied asymptotically for any infinitely small ε. Since the coefficients of each power of ε are independent of the parameter itself, each coefficient must be identically zero in order to satisfy the equation for any arbitrarily small ε.

[17] For small ε, terms of $O(\varepsilon)$ become irrelevant. Only terms of order $O(1)$, $O(\varepsilon^{-1})$, $O(\varepsilon^{-2})$ have to be considered to determine the terms up to the first order in expansion (12). Thus we obtain the equations

$$\partial_{\hat{t}} \hat{C}_0 + \text{Pe}\, \hat{\boldsymbol{u}}^*|_{\hat{x}} \cdot \nabla_{\hat{x}} \hat{C}_0 - \nabla_{\hat{x}}^2 \hat{C}_0 + \text{Pe}\, \hat{\tilde{\boldsymbol{u}}}\Big|_{\hat{y}} \cdot \nabla_{\hat{x}} \hat{C}_1 + \text{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{C}_2 - \nabla_{\hat{y}}^2 \hat{C}_2 - 2 \nabla_{\hat{x}} \cdot \nabla_{\hat{y}} \hat{C}_1 = 0 \tag{13}$$

$$\text{Pe}\, \hat{\tilde{\boldsymbol{u}}}\Big|_{\hat{y}} \cdot \nabla_{\hat{x}} \hat{C}_0 + \text{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{C}_1 - \nabla_{\hat{y}}^2 \hat{C}_1 - 2 \nabla_{\hat{x}} \cdot \nabla_{\hat{y}} \hat{C}_0 = 0 \tag{14}$$

$$\text{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{C}_0 - \nabla_{\hat{y}}^2 \hat{C}_0 = 0 \tag{15}$$

We dropped the superscript Pe to simplify the notation. Since problems at the mesoscopic scale can be regarded as unbounded, the unique solution of the local equation (15) is constant at mesoscopic scales and depends only on the macroscopic variable $\hat{\boldsymbol{x}}$, i.e.

$$\hat{C}_0 = \hat{C}_0(\hat{\boldsymbol{x}}, t). \tag{16}$$

If we define a vector $\chi(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}) = \ell\, \hat{\chi}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$ such that

$$\hat{C}_1(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{\boldsymbol{t}}) = -\hat{\chi}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}) \cdot \nabla_{\hat{x}} \hat{C}_0(\hat{\boldsymbol{x}}, \hat{\boldsymbol{t}}), \tag{17}$$

one can easily rewrite equation (14) in the form

$$\text{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{\chi}_k - \nabla_{\hat{y}}^2 \hat{\chi}_k = \text{Pe}\, \hat{\tilde{u}}_k\Big|_{\hat{y}}, \tag{18}$$

as the smooth macroscopic gradient $\nabla_{\hat{x}} \hat{C}_0(\hat{\boldsymbol{x}}, \hat{t})$ can be regarded as constant in the local problem. The last term of equation (14) vanishes due to (16). Vector χ has the dimension of a length and represents the part of a particle trajectory fluctuating around the mean trajectory in the asymptotic limit (steady state behavior). Finally, we average the macroscopic equation (13) and using the property of the velocity to be divergence free, we can write

$$\partial_{\hat{t}} \hat{C}_0 + \text{Pe}\, \hat{\boldsymbol{u}}^*|_{\hat{x}} \cdot \nabla_{\hat{x}} \hat{C}_0 - \nabla_{\hat{x}}^2 \hat{C}_0 + \nabla_{\hat{x}} \cdot \left\langle \hat{C}_1 \hat{\tilde{\boldsymbol{u}}}\Big|_{\hat{y}} \right\rangle + \left\langle \nabla_{\hat{y}} \cdot \left(\hat{C}_2 \hat{\boldsymbol{u}}\Big|_{\hat{y}} \right) \right\rangle - \left\langle \nabla_{\hat{y}}^2 \hat{C}_2 \right\rangle - 2 \nabla_{\hat{x}} \cdot \left\langle \nabla_{\hat{y}} \hat{C}_1 \right\rangle = 0. \tag{19}$$

Since the solution is a stationary random process in the variable $\hat{\boldsymbol{y}}$, the last three terms of equation (19) vanish, and using definition (17) we write

$$\partial_{\hat{t}} \hat{C}_0 + \text{Pe}\, \hat{\boldsymbol{u}}^*|_{\hat{x}} \cdot \nabla_{\hat{x}} \hat{C}_0 - \nabla_{\hat{x}} \cdot \left[\left(\mathbf{I} + \delta \hat{\mathbf{D}}^M \right) \nabla_{\hat{x}} \hat{C}_0 \right] = 0, \tag{20}$$

where

$$\delta \hat{\mathbf{D}}^M := \text{Pe} \left\langle \hat{\chi} \otimes \hat{\tilde{\boldsymbol{u}}}\Big|_{\hat{y}} \right\rangle \tag{21}$$

is the dimensionless heterogeneity-induced part of the macrodispersion tensor in the asymptotic limit $\varepsilon \ll 1$. The symbol $\otimes$ denotes the tensor product of two vectors yielding a tensor with components $\delta \hat{D}_{ik}^M := \text{Pe} \langle \hat{\chi}_i \hat{\tilde{u}}_k |_{\hat{y}} \rangle$.

[18] The upscaled equation (20) with the additional local equation (18) and appropriate boundary and initial conditions, determines the unique solution of the problem (8) in the asymptotic limit $\varepsilon \ll 1$. We assumed that the velocity field is divergence-free. No additional hypothesis has been made either on the equation governing the flow field or on the relation between mean drift and zero-mean residual. Therefore the results of the previous section apply to any transport problem in which the velocity field exhibits a multiple-scale behavior with well-separated scales. It is interesting to notice that our upscaled and additional equations coincide with those obtained by *Brenner* [1980] in studying Taylor dispersion. The equivalence between Brenner's method of moments and multiple-scale analysis was pointed out in the past by *Rubinstein and Mauri* [1986], *Mei* [1992], and *Auriault and Adler* [1995], but they have to introduce ad hoc assumptions to ensure this equivalence that is never complete. We overcame these problems by using the kinematic relationship (6), which avoids mixing terms of a different order or separate term of the same order. In the following we will focus on macroscopic dispersion in laminar flow through porous media. Assuming the flow governed by Darcy's law, we look for a relation between mean drift and random component of the velocity. This introduces some simplification and helps us to discuss the properties of the dispersion tensor that is the final scope of our paper.

4. Asymptotic Macrodispersivity in Porous Media (Wide-Scale Separation)

4.1. Homogenization of the Flow Problem

[19] The homogenization of an elliptic equation is a classical issue of the multiple-scale analysis in heterogeneous media. It has been rigorously treated in periodic media by *Bensoussan et al.* [1978] and *Sanchéz-Palencia* [1980], and in the stochastic case by *Papanicolaou and Varadhan* [1981]. For simplicity, *Papanicolaou and Varadhan* [1981] consider a stationary hydraulic conductivity tensor $\mathbf{k}$, $k_{ij} = k_{ij}(\boldsymbol{y})$, and a smooth source term, but extensions to more general cases are straightforward. In particular, one can easily extend to stochastic media the results for periodic media obtained by *Bensoussan et al.* [1978] and *Sanchéz-Palencia* [1980] and assume that hydraulic conductivity and the source term are two-scale functions, i.e., $k_{ij} = k_{ij}(\boldsymbol{x}, \boldsymbol{y})$ and $q = q(x, y)$. Following *Sanchéz-Palencia* [1980] and using the steps of section 3, we expand the piezometric head determined by

$$\nabla_\xi \cdot \left(\mathbf{k} \nabla_\xi h\right) = -q \tag{22}$$

in a power series in terms of ε, i.e., $h = h_0 + \varepsilon\, h_1 + \varepsilon^2 h_2 + \ldots$. It is straightforward to show that the zero-order dimensionless velocity is

$$\hat{\boldsymbol{u}}_{\boldsymbol{0}} = -\left[\hat{\mathbf{k}}\left(\mathbf{I} + \nabla_{\hat{y}} \otimes \hat{\boldsymbol{w}}\right)\right] \nabla_{\hat{x}}\, \hat{h}_0, \tag{23}$$

where $\left(\nabla_{\hat{y}} \otimes \hat{\boldsymbol{w}}\right)_{ij} = \partial_{\hat{y}_i} \hat{w}_k \cdot \hat{\boldsymbol{w}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$ solves the local equation in the mesoscale variable

$$\nabla_{\hat{y}} \cdot \left(\hat{\mathbf{k}} \nabla_{\hat{y}} \hat{w}_k\right) = -\left(\nabla_{\hat{y}} \cdot \hat{\mathbf{k}}\right)_k, \tag{24}$$

and $\hat{h}_0$ is the solution of the large-scale equation

$$\nabla_{\hat{x}} \cdot \left(\left\langle \hat{\mathbf{K}} \right\rangle \nabla_{\hat{x}} \hat{h}_0\right) = -\hat{q}^*. \tag{25}$$

where $\hat{\mathbf{K}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}) := \left[\hat{\mathbf{k}}\left(\mathbf{I} + \nabla_{\hat{y}} \otimes \hat{\boldsymbol{w}}\right)\right]$ and $\hat{q}^* = \hat{q}^*(\hat{\boldsymbol{x}})$ is a smoothed source term. Note that the velocity $\hat{\boldsymbol{u}}_0$ depends on the mesoscopic variable $\hat{\boldsymbol{y}}$ before the spatial average has been performed. The tensor $\hat{\mathbf{K}}$ depends only on the hydraulic conductivity $\hat{\mathbf{k}}$, which is a medium property and is independent of flow configuration.

[20] The mean drift is $\hat{\boldsymbol{u}}^*(\hat{\boldsymbol{x}}) = -\left\langle \hat{\mathbf{K}} \right\rangle \nabla_{\hat{x}} \hat{h}_0(\hat{\boldsymbol{x}})$ and the zero-mean residual $\hat{\tilde{\boldsymbol{u}}} = \hat{\tilde{\boldsymbol{u}}}_0 + \hat{\boldsymbol{u}}_1 \varepsilon + \hat{\boldsymbol{u}}_2 \varepsilon^2 + \ldots$, where the zero order coefficient is $\hat{\tilde{\boldsymbol{u}}}_0 = \hat{\boldsymbol{u}}_0 - \hat{\boldsymbol{u}}_0^*$. If we define a tensor

$$\tilde{\mathbf{V}} := -\hat{\mathbf{K}} \left\langle \hat{\mathbf{K}} \right\rangle^{-1} + \mathbf{I}, \tag{26}$$

where $\langle \hat{\mathbf{K}} \rangle^{-1}$ is the inverse tensor of $\langle \hat{\mathbf{K}} \rangle$ such that $\langle \hat{\mathbf{K}} \rangle \langle \hat{\mathbf{K}} \rangle^{-1} = \mathbf{I}$, the definition of $\langle \hat{\mathbf{K}} \rangle$ with equations (22) and (24) yields

$$\hat{\tilde{\boldsymbol{u}}}_0 = -\hat{\mathbf{K}} \nabla_{\hat{x}} \hat{h}_0 - \hat{\boldsymbol{u}}^* = -\left[\hat{\mathbf{K}} \left\langle \hat{\mathbf{K}} \right\rangle^{-1} - \mathbf{I}\right] \left\langle \hat{\mathbf{K}} \right\rangle \nabla_{\hat{x}} \hat{h}_0 = \tilde{\mathbf{V}} \hat{\boldsymbol{u}}^*. \tag{27}$$

The inverse of $\langle \hat{\mathbf{K}} \rangle$ always exists because the tensor is positive definite. The tensor $\tilde{\mathbf{V}}$ depends only on medium properties and not on the flow configuration. The tensor $\hat{\mathbf{K}} \langle \hat{\mathbf{K}} \rangle^{-1}$ transforms the macroscopic mean drift into the local mesoscopic velocity. It is analogous to the microscopic local tensor of the porous medium introduced by *Nikolaevskii* [1959] that maps the Darcy velocity into the local velocity inside the pores.

4.2. Homogenization of the Transport Equation in Porous Media

[21] Now we can insert $\hat{\tilde{\boldsymbol{u}}} = \hat{\tilde{\boldsymbol{u}}}_0 + \hat{\boldsymbol{u}}_1 \varepsilon + \hat{\boldsymbol{u}}_2 \varepsilon^2 + \ldots$ in the transport equation (10) and perform an analysis analogous to the one shown in section (3). It is easy to prove that all terms of order higher than one in the expansion do not contribute to the final equations and the result is still given by equations (18), (20), and (21). Substituting $\hat{\tilde{\boldsymbol{u}}} = \tilde{\mathbf{V}} \hat{\boldsymbol{u}}^*$ and $\hat{\mathbf{u}} = (\tilde{\mathbf{V}} + \mathbf{I})\hat{\mathbf{u}}^*$ in equations (18) and (21), we can write the components of the macrodispersion tensor in (20) as $\delta \hat{D}^M_{ik} = \mathrm{Pe} \left\langle \hat{\chi}_i \tilde{V}_{jk} \right\rangle \hat{u}_j^* \big|_{\hat{y}}$, where the components of $\hat{\boldsymbol{\chi}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$ now solve the equation

$$\mathrm{Pe}\left[\left(\tilde{\mathbf{V}} + \mathbf{I}\right)\hat{\boldsymbol{u}}^* \cdot \nabla_{\hat{y}} \hat{\chi}_k - \left(\tilde{\mathbf{V}} \hat{\boldsymbol{u}}^*\right)_k\right] = -\nabla^2_{\hat{y}} \hat{\chi}_k. \tag{28}$$

In the following we focus on advectively dominated transport, namely $\mathrm{Pe} \gg 1$, which is the most interesting case for practical applications, and we neglect the diffusive term on the right hand side of (28). Notice that in this case the vector χ is independent of the Peclet number. Recalling that $\boldsymbol{u}^* = (UL/\ell)\hat{\mathbf{u}}^*|_{\hat{y}}$, $\boldsymbol{\chi} = \ell\, \hat{\boldsymbol{\chi}}$, and $\boldsymbol{y} = \ell\, \hat{\boldsymbol{y}}$, we write the dimensional dispersion coefficients as

$$\delta D^M_{ik} = \left\langle \chi_i \tilde{V}_{jk} \right\rangle u_j^*, \tag{29}$$

and equation (28) in the form

$$\left[\left(\tilde{\mathbf{V}} + \mathbf{I}\right)\frac{\boldsymbol{u}^*}{u^*} \cdot \nabla_{\hat{y}} \chi_k - \left(\frac{\tilde{\mathbf{V}} \boldsymbol{u}^*}{u^*}\right)_k\right] = 0, \tag{30}$$

where $u^* := (\boldsymbol{u}^* \cdot \boldsymbol{u}^*)^{1/2}$ is the absolute value of the velocity. Equation (30) demonstrates that the macrodispersion coefficients (29) depend linearly on the absolute value of the mean drift velocity, χ being independent of u^*.

[22] Since $\langle \chi \otimes \tilde{\mathbf{V}} \rangle_{ijk} = \langle \chi_i \tilde{V}_{jk} \rangle$ is a third-rank tensor, it is never isotropic and it is not able to describe the dispersivity of an isotropic medium. This suggests that the third-rank tensor is actually the product between the direction of mean drift and a fourth-rank tensor, which represents a property of the medium independent of the flow. To prove this statement, we introduce the Green"s function that solves $(\tilde{\mathbf{V}} + \mathbf{I})(\boldsymbol{u}^*/u^*) \cdot \nabla_{\hat{y}} G = \delta(y = y')$; it is independent of the absolute value of the velocity and depends only on its direction $\hat{\boldsymbol{e}}^* = \boldsymbol{u}^*/u^*$, which is a function of the megascopic variable $\boldsymbol{x}$. Solution of equation (30) is

$$\chi(\boldsymbol{x}, \boldsymbol{y}) = -\int G(\hat{\boldsymbol{e}}^*, \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{y}') \tilde{\mathbf{V}}(\boldsymbol{x}, \boldsymbol{y}') \frac{\boldsymbol{u}^*(\boldsymbol{x})}{u^*(\boldsymbol{x})} \mathrm{d}\boldsymbol{y}'. \quad (31)$$

Inserting equation (31) into (29) we obtain

$$\delta D_{ik}^M = \alpha_{ilkj} \frac{u_l^* u_j^*}{u^*}, \quad (32)$$

where we have defined a macrodispersivity as a fourth-rank tensor with components

$$\begin{aligned} \alpha_{ilkj} &:= \left\langle \int \tilde{\mathbf{V}}' \otimes \tilde{\mathbf{V}}\ G\ \mathrm{d}\boldsymbol{y}' \right\rangle_{ilkj} \\ &= \left\langle \int \tilde{V}_{il}(\boldsymbol{x}, \boldsymbol{y}') \tilde{V}_{kj}(\boldsymbol{x}, \boldsymbol{y})\ G(\hat{\boldsymbol{e}}^*, \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{y}') \mathrm{d}\boldsymbol{y}' \right\rangle. \end{aligned} \quad (33)$$

From the definition of the averaging operator (2) we conclude that the fourth-rank tensor is symmetric, $\alpha_{ilkj} = \alpha_{ijkl}$, but in general not isotropic. The Green"s function depends on the mean flow direction and the macrodispersivity (33) depends on the angle between the mean drift and the principal axes of the heterogeneous structure of the medium. If the medium is isotropic at the mesoscale and statistically isotropic at macroscale, the macrodispersion tensor becomes diagonal in a coordinate system along the streamlines, but its entries are not isotropic. On the other hand, the macrodispersivity becomes isotropic since the result of the integral (33) becomes independent of the flow direction. Finally, we observe that (33) is determined by local equations in the mesoscopic variable y. Any quantity depending on x can be considered as a constant at the local scale (see section 2) and appears as a parameter: the flow is locally uniform. This is a straightforward consequence of the scale separation and demonstrates the very intuitive and physically based observation that the flow can be regarded as locally uniform if it is observed at a sufficiently small scale, i.e., if variations take place at a spatial scale large enough compared to the local scale.

4.3. An Explicit Result: Macrodispersivity in Lowest-Order Perturbation Theory

[23] Although the validity of (33) is not limited by the amplitude of velocity fluctuations in the asymptotic limit $\varepsilon \ll 1$, analytical results are difficult to obtain apart from very simple cases and the equation can be solved only numerically. In this section we restrict our attention to weakly fluctuating velocity fields. This allows us to obtain explicit results to be compared with those obtained by other techniques in uniform flow fields [e.g., *Gelhar and Axness*, 1983; *Dagan*, 1984; *Kitanidis*, 1988; *Indelman and Dagan*, 1999].

[24] If the zero-mean residual of the velocity is small, we can write the equation that defines the Green"s function associated to the local problem as

$$\frac{\boldsymbol{u}^*(\boldsymbol{x}) + \tilde{\boldsymbol{u}}(\boldsymbol{x}, \boldsymbol{y})}{u^*(\boldsymbol{x})} \cdot \nabla_y G \approx \hat{\boldsymbol{e}}^*(\boldsymbol{x}) \cdot \nabla_y G = \delta(\boldsymbol{y} - \boldsymbol{y}'). \quad (34)$$

Let us indicate with η_1 the coordinate parallel and with η_2 and η_3 the coordinates orthogonal to the mean flow. In this reference system the solution of (34) takes a very simple form:

$$G(\boldsymbol{\eta}, \boldsymbol{\eta}') = \Theta(\eta_1 - \eta_1')\delta(\eta_2 - \eta_2')\delta(\eta_3 - \eta_3'), \quad (35)$$

where Θ is the step function. The Green"s function (35) can be understood as the trajectory, in the lowest-order approximation, of a particle that is released at the location $\boldsymbol{\eta}'$. Inserting (35) into (32) and (33), we obtain in the new reference system

$$\begin{aligned} \delta D_{\eta_i\eta_k}^M &= u^* \int_{\eta_1' < \eta_1} \left\langle \frac{\tilde{u}_{\eta_i}(\eta_1', \eta_2, \eta_3)\tilde{u}_{\eta_k}(\eta_1, \eta_2, \eta_3)}{u^* u^*} \right\rangle \mathrm{d}\eta_1' \\ &= u^* \int_{\eta_1' < \eta_1} R_{\eta_i\eta_k}(\eta_1', \eta_1)\mathrm{d}\eta_1', \end{aligned} \quad (36)$$

where we performed the spatial integration with respect to the transverse coordinates η_2, η_3 and employed the assumption of scale separation. We also used $u_\eta^* := (u_{\eta_i}^* u_{\eta_i}^*)^{1/2} = u^*$. $R_{\eta_i\eta_k}(\eta_1', \eta_1)$ is the dimensionless velocity correlation function for an increment in the mean flow direction. It is determined by the statistics of the conductivity field via Darcy’s law and is a function of $\boldsymbol{x}$, since the flow direction is $\hat{\boldsymbol{e}}^* = \hat{\boldsymbol{\eta}}_1$. In a uniform flow field, $\hat{\boldsymbol{\eta}}_1$ constant and the macrodispersion coefficients (36) reduce to the formulae known since the pioneering work of *Gelhar and Axness* [1983] and *Dagan* [1984]. Indeed, the dimensionless velocity correlation function becomes translation-invariant and for a mean flow in the x_1 -direction the macrodispersive coefficient reduces to

$$\delta D_{ik}^M = u^* \int_{-\infty}^{x_1} R_{ik}(x_1 - x_1')\mathrm{d}x_1' = u^* \int_0^{\infty} R_{ik}(x_1')\mathrm{d}x_1'. \quad (37)$$

In arbitrary nonuniform flow fields only the reference system of coordinates is different, since the absolute value of the mean velocity does not enter into equation (34). The tensor $\delta D_{\eta_i\eta_k}^M$ is in general nondiagonal, since its principal axes coincide neither with the flow direction, nor with the principal axes of anisotropy of the medium. Its components depend on the angle between the mean flow and the principal axis of anisotropy of the medium, which is space dependent. Nevertheless, due to the scale separation the local problem in the variable y is still uniform, and locally (x appears as a

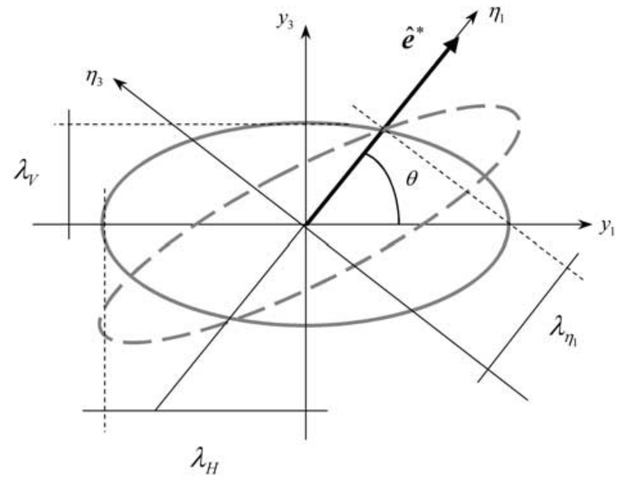

Figure 2. Anisotropy ellipse of the log conductivity (solid shaded line), and of the dispersion (dashed shaded line) with the mean flow direction $\hat{\boldsymbol{e}}^*$ forming an angle θ with the bedding plane y_1.

parameter) equation (36) reduces to the uniform flow case with arbitrary orientation of the anisotropy tensor [see *Gelhar and Axness*, 1983; *Dagan*, 1984].

[25] To illustrate these results we focus on the longitudinal dispersion coefficient, $\delta D_L^M := \delta D_{\eta_1\eta_1}^M$. We assume a classical lognormal conductivity distribution $\mathbf{k}(\boldsymbol{x}, \boldsymbol{y}) = k^*(\boldsymbol{x}) \exp[\tilde{f}(\mathbf{y})]\mathbf{I}$, where $k^*(\mathbf{x})\mathbf{I} = \exp\langle \ln \mathbf{k}(\boldsymbol{x}, \boldsymbol{y})\rangle$ is the geometric mean and $\tilde{f}$ is a zero-mean normally distributed random process with a Gaussian correlation function

$$\langle \tilde{f}(\boldsymbol{y}')\tilde{f}(\boldsymbol{y})\rangle = \exp\left[-\frac{(y_1 - y_1')^2}{\lambda_H^2} - \frac{(y_2 - y_2')^2}{\lambda_H^2} - \frac{(y_3 - y_3')^2}{\lambda_V^2}\right], \tag{38}$$

which corresponds to an anisotropic medium with correlation length λ_H in the bedding plane and λ_V in the orthogonal direction (see Figure 2). The velocity correlation function can be written as $R_{\eta_i\eta_k}(\eta_1', \eta_1) = P_{\eta_i\eta_k}\langle \tilde{f}(\eta_1', \eta_2, \eta_3)\tilde{f}(\eta_1, \eta_2, \eta_3)\rangle$, where the operator $P_{\eta_i\eta_k}$ ensures a divergence free velocity field and relates the velocity and the conductivity correlation functions. As mentioned above, the mean flow direction $\hat{\boldsymbol{e}}^*$ can be regarded as constant at the local scale and forms an angle θ (Figure 2) with the bedding. This angle is not constant at the macroscale but depends on the macroscopic variable, $\theta = \theta(\boldsymbol{x})$ (Figure 3); the local situation at each point of Figure 3 is described by a picture analogous to Figure 2.

[26] Since the mean drift is locally uniform, the evaluation of the integral (36) to compute the longitudinal macrodispersion coefficient is straightforward and yields

$$\delta D_L(\boldsymbol{x}) = \sigma_f^2 I_{\eta_1}(\boldsymbol{x}) u * (\boldsymbol{x}) \tag{39}$$

or for the dispersivity $\alpha_L(\boldsymbol{x}) := \alpha_{\eta_1\eta_1\eta_1\eta_1}(\boldsymbol{x}) = \sigma_f^2 I_{\eta_1}(\boldsymbol{x})$, where σ_f^2 is the variance of the log conductivity field and $I_{\eta_1} = (\sqrt{\pi}/2)\lambda_{\eta_1}$ the integral scale in the direction $\hat{\boldsymbol{\eta}}_1\hat{\boldsymbol{e}}$ (Figure 2). The correlation length in the mean flow direction is

$$\lambda_{\eta_1}(\boldsymbol{x}) = \frac{1}{\sqrt{\frac{\cos^2\theta(\boldsymbol{x})}{\lambda_H^2} + \frac{\sin^2\theta(\boldsymbol{x})}{\lambda_V^2}}}. \tag{40}$$

If the flow is essentially horizontal, $\theta = 0, \alpha_L = (\sqrt{\pi}/2)\sigma_f^2\lambda_H$, which is equivalent to the asymptotic longitudinal dispersivity found in uniform and in radial transport situations (for further references see, e.g., *Indelman and Dagan* [1999] and *Attinger et al.* [2001]). An analogous result holds if $\lambda_V = \lambda_H$, showing that the large-scale dispersivity in isotropic media describes a medium property and therefore does not depend on the flow configuration. For almost vertical flow, $\theta = \pi/2, \alpha_L = (\sqrt{\pi}/2)\sigma_f^2\lambda_V$. The full behavior of the longitudinal dispersivity for different angles is plotted in Figure 4 for different values of the anisotropy ratio λ_V/λ_H .

5. Extension to Preasymptotic Transport Behavior

[27] In the previous sections our analysis is based on the assumption that the two scales involved are widely separated. However, the limit $\varepsilon \to 0$ cannot be performed strictly at early times or in many practical applications, in which an intermediate scale prevents the solution from reaching its asymptotic form. In this section we generalize our two-scale expansion analysis to finite ε. Including terms of higher order in ε is unlikely to give the correct qualitative behavior if the scales are not well separated because our expansion is not a simple Taylor expansion, but an asymptotic expansion. Thus we describe this behavior via a transient additional equation and the introduction of a cutoff value set by the observation scale.

[28] The finiteness of ε has important consequences on the relationships between the two variables defined by $d\hat{\boldsymbol{y}} = d\hat{\boldsymbol{x}}/\varepsilon$ (see section 2). Analogously, to section 2 we can split a

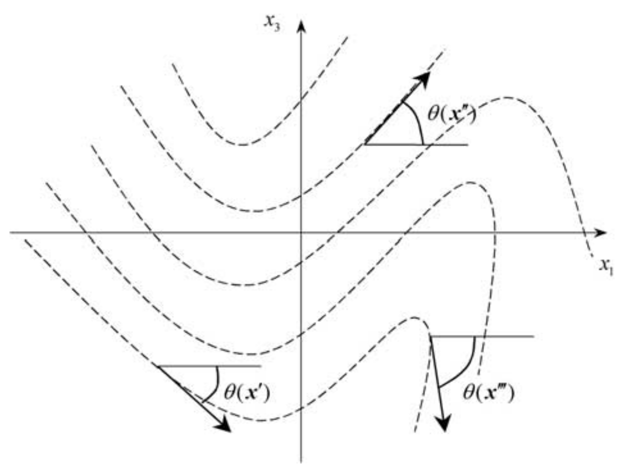

Figure 3. Streamlines for a nonuniform mean flow (dashed lines). The mean flow directions $\hat{\boldsymbol{e}}^*$ (arrows) form with the bedding plane x_1 an angle θ, which is a function of the large-scale variable $\boldsymbol{x}$. The local situation at each point $\boldsymbol{x}$ is described in Figure 2.

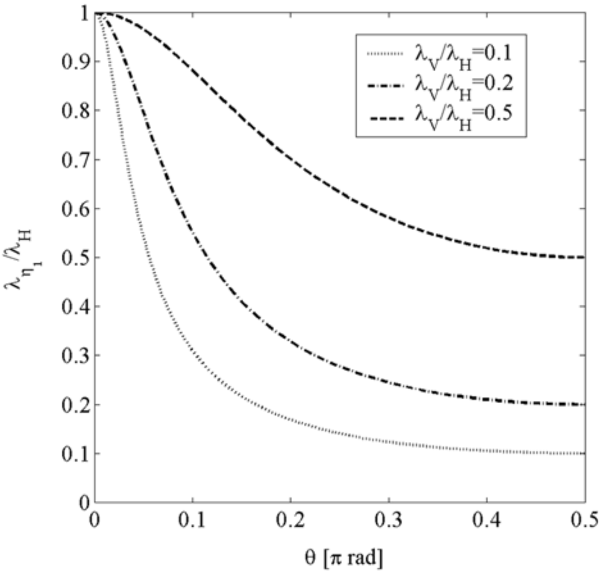

Figure 4. Plot of $\lambda_{\eta_1}/\lambda_H$ as a function of $\theta \in [0, \pi/2]$ for different values of the anisotropy ratio λ_V/λ_H.

two-scale function into a smoothed part and a zero-mean fluctuation, i.e., $\psi = \langle\psi\rangle^{\varepsilon} + \tilde{\psi}^{\varepsilon}$, where the superscript $^{\varepsilon}$ indicates that now, in contrast to the asymptotic limit result ($\varepsilon \ll 1$), both the mean value and the fluctuation depend on the size of the averaging volume, thus on ε. The splitting corresponds to a filtering procedure that filters out small-scale fluctuations up to a certain cutoff length scale determined by the size of the averaging volume. This cutoff length is set by L, the resolution scale of the spatially nonuniform macroscopic flow field, which has the same order of magnitude as ℓ, because ε is now finite. The spatial average $\langle\ \rangle^{\varepsilon}$ is not equal to the ensemble average due to lack of ergodicity with respect to the mesoscopic fluctuations such that the behavior of a single realization differs from the ensemble behavior. With this in mind, we split the velocity field into $\hat{\boldsymbol{u}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}) = \langle\hat{\boldsymbol{u}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})\rangle^{\varepsilon} + \hat{\tilde{\boldsymbol{u}}}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})$ and we proceed rescaling as in section 2. We obtain an equation analogous to (11), whose solution we expand in the form

$$\begin{aligned}\hat{C}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}, \hat{t}^{\ell}) &= \left\langle \hat{C}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}, \hat{t}^{\ell}) \right\rangle^{\varepsilon} + \varepsilon \hat{\tilde{C}}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}, \hat{t}^{\ell}) \\ &= \hat{C}_0^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{t}, \hat{t}^{\ell}) + \varepsilon \hat{\tilde{C}}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}, \hat{t}^{\ell}).\end{aligned} \tag{41}$$

Terms of order higher than $O(\varepsilon)$ are implicitly considered in $\hat{\tilde{C}}^{\varepsilon}$ and the dependence on $\hat{t}^{\ell}$ is maintained: because of the finiteness of ε, the solution may yet not have become steady state with respect to mesoscopic timescale. As in section 3, we collect terms of the same power of ε and require that coefficients of order $O(\varepsilon^{-1})$, $O(1)$ are identically zero. Thus we obtain

$$\partial_{\hat{t}} \hat{C}_0^{\varepsilon} + \mathrm{Pe}\langle\hat{\boldsymbol{u}}\rangle^{\varepsilon}|_{\hat{x}} \cdot \nabla_{\hat{x}} \hat{C}_0^{\varepsilon} - \nabla_{\hat{x}}^2 \hat{C}_0^{\varepsilon} + \mathrm{Pe}\, \hat{\tilde{\boldsymbol{u}}}^{\varepsilon}\Big|_{\hat{y}} \cdot \nabla_{\hat{x}} \hat{\tilde{C}}^{\varepsilon} = 2\nabla_{\hat{x}} \cdot \nabla_{\hat{y}} \hat{\tilde{C}}^{\varepsilon}, \tag{42}$$

$$\partial_{\hat{t}^{\ell}} \hat{\tilde{C}}^{\varepsilon} + \mathrm{Pe}\, \hat{\tilde{\boldsymbol{u}}}^{\varepsilon}|_{\hat{y}} \cdot \nabla_{\hat{x}} \hat{C}_0^{\varepsilon} + \mathrm{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{\tilde{C}}^{\varepsilon} - \nabla_{\hat{y}}^2 \hat{\tilde{C}}^{\varepsilon} = 0. \tag{43}$$

Whereas in section 3 the coefficients of the expanded solution are independent of ε and this passage is rigorous, here its feasibility is postulated. This introduces an approximation.

[29] In order to decouple equations (42) and (43) we define a vector $\chi(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}^{\ell}) = \ell\hat{\chi}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}^{\ell})$ such that

$$\hat{\tilde{C}}_{\varepsilon}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}, \hat{t}^{\ell}) = -\hat{\chi}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}, \hat{t}^{\ell}) \cdot \nabla_{\hat{x}} \hat{C}_0^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{t}, \hat{t}^{\ell}). \tag{44}$$

Inserting equation (44) into (42) and (43) and averaging the macroscopic equation we obtain

$$\begin{aligned}&\partial_{\hat{t}} \hat{C}_0^{\varepsilon} + \mathrm{Pe}\langle\hat{\boldsymbol{u}}\rangle^{\varepsilon}|_{\hat{x}} \cdot \nabla_{\hat{x}} \hat{C}_0^{\varepsilon} - \nabla_{\hat{x}} \cdot \left[\left(\mathbf{I} + \boldsymbol{\delta}\hat{\mathbf{D}}^{\varepsilon}\right) \nabla_{\hat{x}} \hat{C}_0^{\varepsilon}\right] \\ &\quad = \left\langle 2\nabla_{\hat{x}} \cdot \nabla_{\hat{y}} \hat{\tilde{C}}^{\varepsilon} \right\rangle^{\varepsilon} = 0,\end{aligned} \tag{45}$$

$$\partial_{\hat{t}^{\ell}} \hat{\chi}_k + \mathrm{Pe}\, \hat{\boldsymbol{u}}|_{\hat{y}} \cdot \nabla_{\hat{y}} \hat{\chi}_k - \nabla_{\hat{y}}^2 \hat{\chi}_k = \mathrm{Pe}\, \hat{\tilde{u}}_k^{\varepsilon}|_{\hat{y}}, \tag{46}$$

where we defined the macroscopic tensor as

$$\boldsymbol{\delta}\hat{\mathbf{D}}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{t}^{\ell}) := \mathrm{Pe}\left\langle \hat{\chi} \otimes \hat{\tilde{\boldsymbol{u}}}^{\varepsilon}\Big|_{\hat{y}} \right\rangle^{\varepsilon}. \tag{47}$$

Equation (44) and the subsequent decoupling correspond to a localization of the problem. The solution of equation (46) determines the macroscopic tensor (47), which appears in the upscaled equation (45). These results show important differences when compared to the asymptotic behavior. First of all, the additional mesoscopic equation (46) is not steady state in the preasymptotic case. Moreover for finite ε the macroscale variable $\hat{\boldsymbol{x}}$ cannot be considered as a parameter; neither on the right hand side of equation (45) nor in the mesoscale problem (46). The latter cannot be considered unbounded, but boundary conditions need to be specified on the boundary of the averaging volume. The solution is a priori statistically nonstationary in $\hat{\boldsymbol{y}}$ and the right hand side of equation (45) does not automatically vanish when averaging. Nevertheless, we can transform the volume integral (averaging operator) into a surface integral and impose appropriate boundary conditions for the mesoscale problem (46) such that this term vanishes. We assume

$$\nabla_x \cdot \int_V \nabla_{\hat{y}} \tilde{C}^{\varepsilon} dy = \nabla_x \cdot \langle\nabla_y \tilde{C}^{\varepsilon}\rangle^{\varepsilon} = \nabla_x \cdot \oint \hat{\boldsymbol{n}} \tilde{C}^{\varepsilon} \approx 0. \tag{48}$$

Thus right hand side in equation (45) is zero and the upscaled equation assumes an advective-dispersive form the important difference with respect to the asymptotic case is that the spatial integral in equation (47) is not performed over an infinite domain, but over a finite support volume $V(\varepsilon)$ and the macrodispersive tensor is now scale dependent (it depends on ε). Moreover, equations (47) and (21) differ in the respective fluctuating velocity fields $\hat{\tilde{\boldsymbol{u}}}^{\varepsilon}$ and $\hat{\tilde{\boldsymbol{u}}}$: $\hat{\tilde{\boldsymbol{u}}}$ is defined as $\hat{\boldsymbol{u}} - \hat{\boldsymbol{u}}^*$ whereas $\hat{\tilde{\boldsymbol{u}}}^{\varepsilon}$ is given by $\hat{\boldsymbol{u}} - \langle\hat{\boldsymbol{u}}(\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}})\rangle^{\varepsilon}$. Therefore the macrodispersion coefficients are independent of the small-scale variable $\hat{\boldsymbol{y}}$ but depend on ε, the ratio between the heterogeneity scale and the observation scale. In the long time limit $\hat{t}^{\ell} \to \infty$, the longitudinal dispersivity can be stated explicitly. For mathematical convenience we replace the spatial average by its ensemble average (mean-field approximation) in the derivation (see Appendix A for details). For isotropic media $\lambda_H = \lambda_V = \lambda$, the longitudinal dispersivity is

$$\alpha_L^{\varepsilon} = \left(\sqrt{\pi}/2\right)\left(1 - \frac{1}{\left(1 + 1/(2\varepsilon^2)\right)^{(d-1)/2}}\right)\lambda\sigma_f^2, \qquad (49)$$

with $\varepsilon = \lambda/L$. The ratio between α_L^{ε} and the asymptotic macrodispersivity, $\alpha_L^{\varepsilon = 0}$, is plotted in Figure 5 both for a two- and a three-dimensional domain. For finite ε , it is always smaller than the asymptotic macrodispersivity because only a finite part of the fluctuations is averaged out, whereas large-scale fluctuations are still resolved and explicitly described in the large-scale flow field, $\langle \hat{\boldsymbol{u}}(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}})\rangle^{\varepsilon} = \hat{\boldsymbol{u}}^* + \langle \hat{\tilde{\boldsymbol{u}}}(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}})\rangle^{\varepsilon} \neq \hat{\boldsymbol{u}}^*$. This implies a fundamental difference to the probabilistic approach: in standard ensemble averaging, all statistical fluctuations of the velocity field within an ensemble of many realizations are averaged out and the ensemble dispersivity is larger than the value given in (49). That demonstrates that standard ensemble averaging does not consistently account for finite scale effects: it tends to overestimate the dispersion coefficient in the single realization.

[30] Finite ε effects are of interest in many practical problems in which a third length scale may prevent the solution from becoming asymptotic. A classical example is given by transport in a dipole flow field where a third length scale is set by the dipole size. In order to model the dipole flow field correctly at macroscopic scale, the observation scale has to be at least of the order of magnitude of the curvature radius of the dipole field. Depending on the dipole length, the observation scale and the heterogeneity scale ℓ may be no longer separated. A numerical test of our theoretical results is beyond the scope of this paper. Numerical simulations will be presented in detail in a following paper (S. Attinger et al., manuscript in preparation, 2002).

6. Summary and Discussion

[31] Most of the approaches to determine the macrodispersion suppose that velocity fluctuations are small compared to the mean flow field. On this basis the transient and asymptotic behavior of a solute cloud can be studied in a probabilistic sense by means of a lowest-order perturbation analysis of the transport problem. Nevertheless, the hypothesis of small fluctuations can be a superfluous restriction, if one is simply interested in the asymptotic behavior and wants to determine the effective parameters to be used in an upscaled advective-dispersive equation that describes the transport process at large time/distance. In this case the small-scale ℓ and the large-scale L are widely separated, $\varepsilon = \ell/L \ll 1$, and a rigorous two scale analysis for small ε can be performed. Using the Homogenization theory approach we derive an upscaled transport equation valid for arbitrary flows and nonstationary media, provided that the heterogeneity scale of the medium is small enough compared to typical lengths of the mean flow. Our analysis of a mesoscale Darcian flow demonstrates that the macrodispersivity is a fourth-rank tensor, α_{ilkj}, which yields a heterogeneity-induced macrodispersion coefficient $\delta D_{ik}^M = \alpha_{ilkj} u_l^* u_j^* / u^*$ in complete analogy with Scheidegger"s tensor, which describes pore scale dispersion. However, the macrodispersivity differs from pore scale dispersivity not only quantitatively but also qualitatively because, on larger scales, soils are statistically anisotropic media with a vertical correlation length that is usually much smaller than the horizontal ones. An important finding is that in the advection dominated case, Pe $\gg$ 1, the asymptotic macrodispersivity is a medium property. If the medium is statistically isotropic, the macrodispersivity is an isotropic tensor; this case formally coincides with the common pore scale dispersivity model. If the medium is statistically anisotropic, the macrodispersivity is an axially symmetric tensor: it depends on the angle θ that the mean flow direction forms with the principal axes of anisotropy. In a uniform flow field, θ is constant and our results reduce to the classical ones for asymptotic macrodispersion found by *Gelhar and Axness* [1983] and *Dagan* [1984]. Nevertheless, homogenization theory enables us to generalized and to study transport phenomena in arbitrary flow configurations because the scale separation yields a separation between the large- and small-scale advective terms. Only the latter contributes to macrodispersion. This case can be treated with difficulty by the Lagrangian approach, because nonuniform mean velocities have an impact on the particle variances that makes identification of dispersivity much more complicated [see, e.g., *Indelman and Dagan*, 1999; *Neuweiler et al.*, 2001]. Therefore only special flow geometries have been considered up to now by this technique: the uniform flow [e.g., *Dagan*, 1984] or the radial flow field [*Indelman and Dagan*, 1999].

[32] In many problems of practical interest a third length scale may prevent the solution from becoming asymptotic (e.g., in a finite domain or in a dipole flow field a third length scale is set by the domain size or by the dipole length, respectively) and the scales are no longer separated. The standard approach to the case of finite ε uses ensemble averaging, but the resulting macrodispersivity is defined in

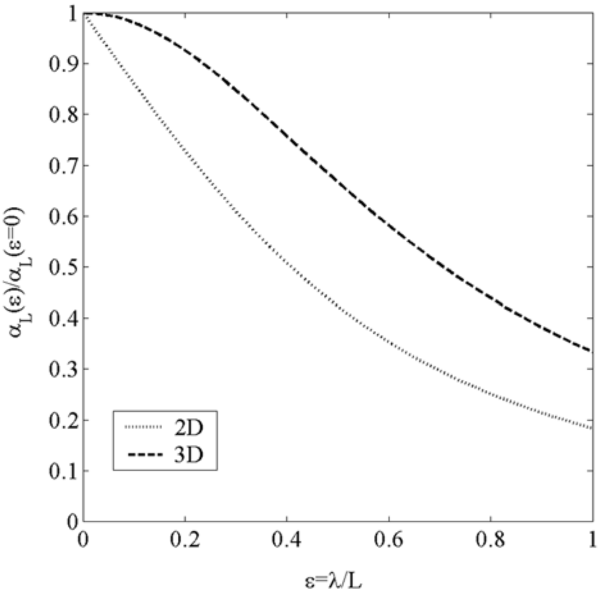

Figure 5. The ratio between the longitudinal dispersivity α_L^{ε} and the asymptotic macrodispersivity, $\alpha_L^{\varepsilon = 0}$, as a function of ε in two and three dimensions.

the ensemble average sense and might not describe the behavior of a single realization because of lack of ergodicity. To overcome this problem we generalize our two-scale analysis to finite ε. Some of the mathematical rigor of the asymptotic case is lost, but these approximate results describe a single realization in contrast to the standard probabilistic approach.

[33] For finite ε, the dispersivity is not a simple medium property, but a scale dependent parameter: it depends on the observation scale (in particular on the size of the averaging volume). It is always smaller than the asymptotic macrodispersion coefficient because the observation scale introduces a cutoff to the part of the velocity fluctuations that are averaged out: fluctuations on a larger scale do not contribute to the dispersion coefficients. For finite ε, the volume average does not equal the ensemble average due to lack of ergodicity and our method differs fundamentally from the probabilistic approach: in standard ensemble averaging, all statistical fluctuations of the velocity field within an ensemble of many realizations are averaged over and the ensemble dispersion coefficients is larger than ours. That demonstrates that standard ensemble averaging does not consistently account for finite scale effects: it tends to overestimate the dispersion coefficient in the single realization.

Appendix A

[34] Here we evaluate $\boldsymbol{\delta}\hat{\boldsymbol{D}}^{\varepsilon}(\hat{\boldsymbol{x}}, \hat{t}^{\ell}) := \mathrm{Pe}\left\langle \hat{\chi} \otimes \hat{\hat{\boldsymbol{u}}}^{\varepsilon}\big|_{\hat{y}} \right\rangle^{\varepsilon}$ in the limit $\hat{t}^{\ell} \to \infty$. The first steps of evaluating the heterogeneity-induced part of dispersion coefficient are the same as in (34)–(36). We obtain

$$\boldsymbol{\delta}\hat{\boldsymbol{D}}^{\varepsilon}_{\eta_1\eta_1}(\hat{\boldsymbol{x}}) = \mathrm{Pe}|\langle\hat{\boldsymbol{u}}\rangle^{\varepsilon}| \int_{\eta_1' < \eta_1} \hat{R}^{\varepsilon}_{\eta_i\eta_k}(\hat{\eta}_1', \hat{\eta}_1)\mathrm{d}\hat{\eta}_1', \tag{A1}$$

which differs from formula (36) in the large flow field $\langle\hat{\mathbf{u}}\rangle^{\varepsilon}$ and in the correlation function $R^{\varepsilon}_{\eta_i\eta_k}$. The small-scale flow field is given by $\hat{\hat{\boldsymbol{u}}}^{\varepsilon} = \hat{\boldsymbol{u}}^{\varepsilon}_{\langle\hat{\boldsymbol{u}}\rangle}$ or

$$\begin{aligned}\hat{\hat{\boldsymbol{u}}}^{\varepsilon} &= \hat{\boldsymbol{u}} - \int_{-1/(2\varepsilon)}^{-1/(2\varepsilon)} d^d\boldsymbol{\xi}'\,\hat{\boldsymbol{u}}(\boldsymbol{\xi}-\boldsymbol{\xi}') = \hat{\boldsymbol{u}} - \int_{-\infty}^{\infty} d^d\boldsymbol{\xi}'\hat{\boldsymbol{u}}(\boldsymbol{\xi}-\boldsymbol{\xi}')\exp\left(-2\varepsilon^2\boldsymbol{\xi}'^2\right)\\ &= \int_{-\infty}^{\infty} d^d\boldsymbol{\xi}'\hat{\boldsymbol{u}}(\boldsymbol{\xi}-\boldsymbol{\xi}')\left(\delta(\boldsymbol{\xi}-\boldsymbol{\xi}') - \exp\left(-2\varepsilon^2\boldsymbol{\xi}'^2\right)\right)\end{aligned} \tag{A2}$$

We get the large-scale flow field $\langle\hat{\boldsymbol{u}}\rangle^{\varepsilon}$ by averaging the flow field $\hat{\boldsymbol{u}}$ over the volume $1/\varepsilon^d$. Moreover, we replaced the sharp volume average by a smoother Gaussian average. Using the translation invariance of the velocity correlations, the correlation function reads

$$\begin{aligned}R^{\varepsilon}_{\eta_i\eta_k}(\boldsymbol{\xi}) = \int_{-\infty}^{\infty} d^d\boldsymbol{\xi}' R_{\eta_i\eta_k}(\boldsymbol{\xi}-\boldsymbol{\xi}')\Bigg(&\delta(\boldsymbol{\xi}-\boldsymbol{\xi}')\\ &-\left(\frac{\varepsilon^2}{2\pi}\right)^{d/2}\exp\left(-2\varepsilon^2\boldsymbol{\xi}'^2\right)\Bigg)\end{aligned} \tag{A3}$$

Inserting $R^{\varepsilon}_{\eta_i\eta_k}$ into (A1) yields for the longitudinal dispersion coefficient after restoring the dimensions,

$$\begin{aligned}\boldsymbol{\delta}\mathbf{D}^{\varepsilon} &= \left|\langle\boldsymbol{u}\rangle^{\varepsilon}\right|\sigma_f^2 I_{\eta_1} - \left|\langle\boldsymbol{u}\rangle^{\varepsilon}\right|\lambda\int^{\eta_1} d\eta_1' \int_{-\infty}^{\infty} d^d\eta'' R_{\eta_i\eta_k}\\ &\quad\cdot(\eta_1-\eta_1'-\eta_1'', -\eta_2'')\left(\frac{\varepsilon^2}{2\pi}\right)^{d/2}\exp\left(-2\varepsilon^2\boldsymbol{\eta}''^2\right)\\ &= \left|\langle\boldsymbol{u}\rangle^{\varepsilon}\right|\sigma_f^2 I_{\eta_1} - \left|\langle\boldsymbol{u}\rangle^{\varepsilon}\right|\lambda\frac{1}{2}\int_{-\infty}^{\infty} d\eta_1' \int_{-\infty}^{\infty} d^d\eta'' R_{\eta_i\eta_k}\\ &\quad\times(\eta_1'-\eta_1'', -\eta_2'')\left(\frac{\varepsilon^2}{2\pi}\right)^{d/2}\exp\left(-2\varepsilon^2\boldsymbol{\eta}''^2\right)\\ &= \left|\langle\boldsymbol{u}\rangle^{\varepsilon}\right|\sigma_f^2 I_{\eta_1} - \left|\langle\boldsymbol{u}\rangle^{\varepsilon}\right|\lambda\sigma_f^2 I_{\eta_1}\int_{-\infty}^{\infty} d\eta_2'' d\eta_3''\\ &\quad\exp\left(\eta_2''^2+\eta_3''^2\right)\left(\frac{\varepsilon^2}{2\pi}\right)^{(d-1)/2}\exp\left(-2\varepsilon^2\left(\eta_2''^2+\eta_3''^2\right)\right)\\ &= |\langle\boldsymbol{u}\rangle^{\varepsilon}|\sigma_f^2 I_{\eta_1}\left(1-\frac{1}{\left(1+1/(2\varepsilon^2)\right)^{(d-1)/2}}\right)\end{aligned}$$

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