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Approximations for transport parameters and self-averaging properties for point-like injections in heterogeneous media

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Abstract

We focus on transport parameters in heterogeneous media with a flow modelled by an ensemble of periodic and Gaussian random fields. The parameters are determined by ensemble averages. We study to what extent these averages represent the behaviour in a single realization. We calculate the centre-of-mass velocity and the dispersion coefficient using approximations based on a perturbative expansion for the transport equation, and on the iterative solution of the Langevin equation. Compared with simulations, the perturbation theory reproduces the numerical results only poorly, whereas the iterative solution yields good results. Using these approximations, we investigate the self-averaging properties. The ensemble average of the velocity characterizes the behaviour of a realization for large times in both ensembles. The dispersion coefficient is not self-averaging in the ensemble of periodic fields. For the Gaussian ensemble the asymptotic dispersion coefficient is self-averaging. For finite times, however, the fluctuations are so large that the average does not represent the behaviour in a single realization.

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1. Introduction

During the last 20 years, stochastic models have become an essential tool for analysing the transport of dissolved pollutants in saturated aquifers. Because of strong spatial variations in the hydraulic conductivity and various other chemical and physical system parameters, the effective large-scale transport properties are different from those found in laboratory-scale experiments. Transport is governed by the heterogeneities of the medium, but due to the lack of knowledge of the detailed local structure in a realistic set-up, the predictive abilities are limited.

In the stochastic approach these heterogeneities are modelled as random, time-independent fields with given statistical properties. The characteristic large-scale behaviour follows from appropriately defined averages over the ensemble of all possible aquifer realizations. The approach has been used to analyse the increase of the dispersion coefficients of a dissolved pollutant in a saturated aquifer owing to local variations of the conductivity, see [14, 13, 5]. A comprehensive overview of the subject can be found in [4, 12]. To show how predictive the stochastic approach for a given realistic set-up is, one has to analyse when the averaged quantities predict the quantities of a single realization. Therefore, it is important to study the self-averaging properties of the transport parameters. This is possible by comparing the transport parameters for a single realization with the ensemble mean, or by the mean square sample-to-sample fluctuations.

In a given aquifer the solute cloud is represented by the concentration field $c(\mathbf{x}, t)$. Its centre-of-mass velocity $u_j(t)$ and the dispersion coefficient $D_{ij}(t)$ are given by

$$u_j(t) = \frac{\mathrm{d}}{\mathrm{d}t} m_j^{(1)}(t) \qquad D_{ij}(t) = \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\left(m_{ij}^{(2)}(t) - m_i^{(1)}(t) m_j^{(1)}(t)\right) \tag{1}$$

where $m_j^{(1)}(t) = \int \mathrm{d}^d x\, x_j c(\mathbf{x}, t)$ and $m_{ij}^{(2)}(t) = \int \mathrm{d}^d x\, x_i x_j c(\mathbf{x}, t)$ are the first two moments of the normalized spatial concentration distribution in d dimensions. For the given aquifer, the observables depend implicitly on the spatial distribution of the heterogeneities. In the stochastic model the medium corresponds to one particular realization of a spatial random process, and the large-scale transport parameters are derived from the ensemble averages. They represent statistic properties of the aquifer ensemble, and therefore, seem to be of limited predictive value with respect to the properties of a single realization. However, for appropriately chosen quantities the fluctuations from realization to realization should become small as soon as the plume has sampled a sufficiently large representative part of the medium. The transport parameters found in different realizations then fluctuate only weakly around the ensemble averages. So, in this case these averages indeed represent 'effective' large-scale parameters characteristic for the single aquifer realization.

Due to (1), the solute is characterized by an effective centre-of-mass velocity $u_j^{\mathrm{eff}}(t) = \overline{u_j(t)}$ and effective dispersion coefficient $D_{ij}^{\mathrm{eff}}(t) = \overline{D_{ij}(t)}$ where the overbar denotes the ensemble average. In the case of an ensemble of a Gaussian random field, the effective transport parameters are known by a perturbation theory analysis [8]. What is still lacking is an analysis of the corresponding sample-to-sample fluctuations as done in [2] for the case of stratified media. The latter is crucial for the reliability of the model.

We investigate the self-averaging properties of the transport parameters by the vehicle of an ensemble of randomly periodic fields. The advantage of such an ensemble is that we can explicitly formulate the local structure of a single realization. Thus, the transport parameters can be derived for a single realization which is done at second order in the flow fluctuations. By this the self-averaging properties are accessible for the periodic ensemble by comparing the values for single realizations with the ensemble mean and by the mean square sample-to-sample fluctuations. As we compare only with the effective transport parameters for the Gaussian ensemble at second order, we do not consider higher orders. Moreover, the statistical properties of the ensemble of periodic fields tend to the properties of the Gaussian random field in the limit of infinite numbers of modes. Hence, we can study the self-averaging properties of the transport parameters by passing to the ensemble of the Gaussian random field for which the direct study by the sample-to-sample fluctuations would be too difficult.

To obtain the transport parameters for a single realization of the periodic ensemble, we present two alternatives: the perturbation theory approach and an approach using a Langevin equation. The effective transport parameters using the perturbative expansion are in good

agreement with numerical simulations, see e.g. [7]. However, use of the perturbation theory for computing the observables for a single realization of the periodic ensemble fails. This will be demonstrated for the case of vanishing diffusion. We bypass this failure using the equivalent description by the Langevin equation and deriving the transport parameters for a single realization by that.

The transport equation and Langevin equation are introduced in section 2, and the perturbation theory approach is summarized in section 3. By iterating the Langevin equation as in section 4, we introduce the new approach to describe the transport behaviour which seems to be a valuable tool. By this we are capable of studying the self-averaging properties of the transport parameters which is done in sections 5 and 6.

2. The model

2.1. Transport equation

The time evolution of a solute in a heterogeneous porous medium is given by an advection–dispersion equation (see, e.g. [4, 13])

$$\frac{\partial}{\partial t}c(\mathbf{x},t) + \nabla \cdot (\mathbf{u}(\mathbf{x})c(\mathbf{x},t)) - \nabla D_0 \nabla c(\mathbf{x},t) = \rho(\mathbf{x})\delta(t) \tag{2}$$

where $c(\mathbf{x}, t)$ is the spatial concentration of the mobile solute. Because of spatial fluctuations in the permeability of the medium, the Darcy velocity $\mathbf{u}(\mathbf{x})$ varies locally. As a consequence of the incompressibility of the fluid, it fulfils $\nabla \cdot \mathbf{u}(\mathbf{x}) = 0$. The tensor D_0 is the local dispersion tensor which includes all the dispersion effects due to fluctuations on microscopic scales. Its general structure is discussed in [19]. As experiments have shown, equation (2) provides a satisfactory description of the mixing process, see [12], and due to the theoretical work in [20] the mixing in miscible flow leads to a flux-induced dispersion which is analogous to Fick's law and results in (2) with a spatially fluctuating D_0 depending on the flow. However, the consideration of this dependence only yields negligible contributions to the transport parameters, see [13, 6]. In the following, we assume the local dispersion tensor to be constant and of diagonal form, $D_{0,ij} = D_0\delta_{ij}$. This kind of model has been successfully used for solute transport in porous media, see [12, 8].

The right-hand side of the transport equation represents the initial condition for an instantaneous injection at $t = 0$. At injection time the total initial concentration is given by $c(\mathbf{x}, t = 0) = \rho(\mathbf{x})$. For the case of a normalized point-like injection, it reduces to a delta function, $\rho(\mathbf{x}) = \delta(\mathbf{x})$. We solely consider transport for a three-dimensional flow field with a point-like injection.

In the stochastic approach the spatially inhomogeneous distribution $\mathbf{u}(\mathbf{x})$ is identified with one single realization of a spatial stochastic process defined by the ensemble of all possible realizations. We assume this process to be statistically translation invariant in space which implies that the ensemble average $\overline{\mathbf{u}(\mathbf{x})}$ does not depend on the spatial position $\mathbf{x}$. We split the fluctuating field into its mean value and the random fluctuations:

$$\mathbf{u}(\mathbf{x}) = \mathbf{u}_0 - \mathbf{w}(\mathbf{x}). \tag{3}$$

The mean $\mathbf{u}_0 = \overline{\mathbf{u}(\mathbf{x})}$ is deterministic and is assumed to be in the 1-direction: $\mathbf{u}_0 = u_0\mathbf{e}_1$. The random fluctuations $w_i(\mathbf{x})$ have zero mean. We presume that they fulfil a Gaussian correlation function $\overline{w_i(\mathbf{x})w_j(\mathbf{x}')}$, which reads in Fourier space:

$$\overline{\tilde{w}_i(\mathbf{k})\tilde{w}_j(\mathbf{k}')} = q_0 u_0^2 (2\pi)^{\frac{3d}{2}} \delta^d(\mathbf{k}+\mathbf{k}') p_i(\mathbf{k}) p_j(\mathbf{k}) \prod_{n=1}^{d} l_n \exp\left(-\frac{k_n^2 l_n^2}{2}\right). \tag{4}$$

The Fourier transformed is defined by $\tilde{g}(\mathbf{k}) = \int_{-\infty}^{\infty} \mathrm{d}^d x\, \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{x}} g(\mathbf{x})$ and $g(\mathbf{x}) = \int_{\mathbf{k}} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{x}} \tilde{g}(\mathbf{k})$, where we use $\int_{\mathbf{k}} \cdots \equiv (2\pi)^{-d} \int \mathrm{d}^d k \cdots$ as a shorthand notation. The parameter q_0 in (4) quantifies the disorder strength, and l_0 denotes the (isotropic) correlation length of the field. The functions $p_i(\mathbf{k})$ are projectors which ensure the incompressibility of the flow fluid. In a d-dimensional system ($d \geqslant 2$), they are given by $p_i(\mathbf{k}) = \delta_{1i} - \frac{k_1 k_i}{\mathbf{k}^2}$, $i = 1, \ldots, d$, see [13, 6]. For the Gaussian random field, all higher correlation functions can be decomposed into products of the given two-point function.

In addition to the ensemble of a Gaussian random field, we consider an ensemble of randomly periodic media which is constructed by a superposition of N cosine modes using randomly chosen parameters. Therefore, the fluctuations $w_i(\mathbf{x})$ of $\mathbf{u}(\mathbf{x})$ are chosen to be, see [17, 9],

$$w_i(\mathbf{x}) = u_0 \sqrt{\frac{2q_0}{N}} \sum_{j=1}^{N} p_i(\mathbf{q}^{(j)}) \cos(\mathbf{q}^{(j)} \cdot \mathbf{x} + \alpha^{(j)}). \tag{5}$$

The Fourier transformation leads to

$$\tilde{w}_i(\mathbf{k}) = u_0 \sqrt{\frac{2q_0}{N}} \frac{(2\pi)^d}{2} \sum_{j=1}^{N} p_i(\mathbf{q}^{(j)}) \left\{ \mathrm{e}^{\mathrm{i}\alpha^{(j)}} \delta^d(\mathbf{k} + \mathbf{q}^{(j)}) + \mathrm{e}^{-\mathrm{i}\alpha^{(j)}} \delta^d(\mathbf{k} - \mathbf{q}^{(j)}) \right\}. \tag{6}$$

Choosing the probability density functions of the parameters appropriately, we can ensure that the ensemble of periodic media has the same statistical properties as the ensemble of the Gaussian field. For this purpose the components of the wave vectors $\mathbf{q}^{(j)}$ are independent and chosen to be of a normal distribution for $j = 1, \ldots, N$, with zero mean and variance l_0^{-2}. The phases $\alpha^{(j)}$ are equally distributed in the interval $[0, 2\pi]$. Thus, the normalized distributions $P_{\mathbf{q}}$ and P_α are given by

$$P_{\mathbf{q}}(\mathbf{q}^{(j)}) = (2\pi)^{-d/2} l_0^d\, \mathrm{e}^{-\frac{1}{2}\mathbf{q}^{(j)2} l_0^2} \qquad P_\alpha(\alpha^{(j)}) = \begin{cases} (2\pi)^{-1} & \text{for } \alpha^{(j)} \in [0, 2\pi] \\ 0 & \text{otherwise.} \end{cases}$$

According to (5), each set of parameters $\{\mathbf{q}^{(j)}, \alpha^{(j)}, N\} := (\mathbf{q}^{(1)}, \ldots, \mathbf{q}^{(N)}, \alpha^{(1)}, \ldots, \alpha^{(N)})$ for fixed N corresponds to a single realization of the flow field. The ensemble average is defined by

$$\begin{aligned} \overline{f(\{\mathbf{q}^{(j)}, \alpha^{(j)}, N\})} = (2\pi)^{-N} & \int_0^{2\pi} \mathrm{d}\alpha^{(1)} \cdots \int_0^{2\pi} \mathrm{d}\alpha^{(N)} \left(\frac{l_0^d}{(2\pi)^{d/2}} \right)^N \\ & \times \int \mathrm{d}^d q^{(1)} \cdots \int \mathrm{d}^d q^{(N)}\, \mathrm{e}^{-\frac{1}{2}\mathbf{q}^{(1)2} l_0^2 - \cdots - \frac{1}{2}\mathbf{q}^{(N)2} l_0^2} f(\{\mathbf{q}^{(j)}, \alpha^{(j)}, N\}). \end{aligned} \tag{7}$$

For the fluctuations $\mathbf{w}(\mathbf{x})$, equation (5), one finds $\overline{\mathbf{w}(\mathbf{x})} = \mathbf{0}$ and $\overline{\tilde{w}_i(\mathbf{k}) \tilde{w}_j(\mathbf{k}')}$ given by (4). So, averages over the ensemble which take into account the correlation upto the order of $O(q_0)$ do not depend on the number of modes N. As shown in [11], higher-order correlation functions exactly show the properties of the Gaussian random field for $N \to \infty$. This proves that the results given by averaging over the ensemble of periodic fields tend to the results of the ensemble of the Gaussian random field in the limit $N \to \infty$, and both kinds of ensembles have the same statistical properties.

This link between both ensembles is important for studying the self-averaging properties of the transport parameters. For the ensemble of randomly periodic fields this can be easily done using the known local structure of the realizations. Then, by increasing the number of modes we can pass to the ensemble of a Gaussian field and investigate the self-averaging properties.

2.2. The Langevin equation

Equivalent to the transport equation is the formulation of transport by the motion of the particles of the solute cloud, which is given by a Langevin equation, see [16]. The transport of a passive tracer particle by a d-dimensional flow field $\mathbf{u}(\mathbf{x})$ is then modelled by the Langevin equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{x}(t) = \mathbf{u}(\mathbf{x}(t)) + \boldsymbol{\xi}(t) \tag{8}$$

where $\mathbf{x}(t)$ denotes the position vector of the particle and $\boldsymbol{\xi}(t)$ is a Gaussian white noise which generates the local diffusion process. It has zero mean $\langle \xi_i(t) \rangle = 0, i = 1, \ldots, d$, and a correlation function $\langle \xi_i(t)\xi_j(t') \rangle = 2D_{ij}\delta(t - t')$, where $\langle \cdot \rangle$ stands for the average over the white noise ensemble. This model can be applied to understand the dispersion of conservative transport in porous media as done e.g. in [2, 9]. However, as shown in [3, 18] results based on this model are often valid only in the long-time limit and do not correctly reproduce the transversal dispersion coefficients which are luckily not the focus here. If the transport is described by the Langevin equation, the moments are given by the solution of (8) averaging the trajectory $x_i(t)$ over all realizations of the ensemble of the white noise, i.e. $m_i^{(1)}(t) = \langle x_i(t) \rangle$ and $m_{ij}^{(2)}(t) = \langle x_i(t)x_j(t) \rangle$. The Gaussian white noise $\xi_j(t)$ is given by a functional distribution $\mathcal{P}[\xi_j(t)] \propto \mathrm{e}^{-(4D_{jj})^{-1}\int_0^t \mathrm{d}t' \xi_j^2(t')}$. The average of $f(\boldsymbol{\xi}(t))$ over the ensemble of the noise is defined by

$$\langle f(\boldsymbol{\xi}(t)) \rangle = \int f(\boldsymbol{\xi}(t)) \prod_{j=1}^{d} \mathcal{D}[\xi_j]\mathcal{P}[\xi_j(t)] \tag{9}$$

with the measure $\mathcal{D}[\xi_j]$. In the case of $D_0 = 0$ the noise $\boldsymbol{\xi}(t)$ vanishes in the Langevin equation.

Note that two types of averages are involved when calculating the effective transport parameters by the Langevin equation: the average over the white noise which generates the local diffusion process, indicated by the brackets, and the average over the disorder ensemble, indicated by the overbar. The order in which these averages are performed is crucial, as discussed e.g. in [2].

2.3. Numerical simulation

For comparison with the theoretical quantities, we compute the centre-of-mass velocity and the dispersion coefficient by numerical simulations. We consider the realizations of the randomly periodic fields and discretize the Langevin equation (8) for a particle tracking simulation. Therefore, we apply an extended Runge–Kutta solver which is described in detail in [10, 7]. The algorithm solves the Langevin equation for one realization of the white noise. To obtain the first two moments of the concentration for a single realization of the randomly periodic field, we average over R realizations of the noise. With the help of the time-discretized and averaged trajectory and velocity, we obtain the numerical centre-of-mass velocity and dispersion coefficient for a given periodic realization. Due to the noise of the numerical dispersion coefficient for small numbers R, we smooth the numerical result of the dispersion coefficient of a single realization for further computations. As shown in [11], by a moving average the curve of $D_{ij}(t)$ is not changed but the noise is efficiently reduced. As the simulations using a large number of realizations of the white noise are very costly, we compute the dispersion coefficient with $10^3 \leqslant R \leqslant 10^4$ and smooth the result in this way. For all numerical simulations we fix $u_0 = 1m/d$, $l_0 = 1m$, and $\tau_u := l_0/u_0 = 1d$.

3. Transport parameters from the perturbative expansion

We summarize the perturbation theory approach for the transport equation and its results for the transport parameters. This approach was successfully applied for deriving the effective transport parameters for the Gaussian random field, see e.g. [1, 8]. Analogously, we apply this approach to obtain the transport properties for a single realization of the ensemble of the randomly periodic fields. However, in this case the perturbative expansion fails to produce good results.

3.1. Perturbation theory approach

We start with the transport equation (2), and the flow field $\mathbf{u}(\mathbf{x})$, given by (3) for a single realization, in the Fourier space:

$$\frac{\partial}{\partial t}\tilde{c}(\mathbf{k},t) + (-\mathrm{i}\mathbf{u}_0\cdot\mathbf{k} + \mathbf{k}D_0\mathbf{k})\tilde{c}(\mathbf{k},t) + \int_{\mathbf{k}'}\mathrm{i}\mathbf{k}\cdot\tilde{\mathbf{w}}(\mathbf{k}')\tilde{c}(\mathbf{k}-\mathbf{k}',t) = \delta(t)\tilde{\rho}(\mathbf{k}). \tag{10}$$

$\tilde{\rho}(\mathbf{k})$ is the Fourier transform of the source term at $t = 0$, which is $\tilde{\rho}(\mathbf{k}) = 1$ for a point-like injection. Using the Fourier transformed concentration $\tilde{c}(\mathbf{k}, t)$, the observables are constructed by

$$u_j(t) = -\mathrm{i}\,\frac{\mathrm{d}}{\mathrm{d}t}\partial_{k_j}(\ln\tilde{c}(\mathbf{k},t))\bigg|_{\mathbf{k}=\mathbf{0}} \qquad D_{ij}(t) = -\frac{1}{2}\,\frac{\mathrm{d}}{\mathrm{d}t}\partial_{k_i}\partial_{k_j}(\ln\tilde{c}(\mathbf{k},t))\bigg|_{\mathbf{k}=\mathbf{0}} \tag{11}$$

where ∂_{k_i} denotes the partial derivative with respect to the $\mathbf{k}$ component in the i direction. We derive a series in the fluctuations $\tilde{\mathbf{w}}$ which is supposed to give an approximation for small $q_0 \ll 1$. Therefore, we transform equation (10) into

$$\tilde{c}(\mathbf{k},t) = \tilde{c}_0(\mathbf{k},t) - \int_{-\infty}^{\infty}\mathrm{d}t'\tilde{c}_0(\mathbf{k},t-t')\int_{\mathbf{k}'}\mathrm{i}\mathbf{k}\cdot\tilde{\mathbf{w}}(\mathbf{k}')\tilde{c}(\mathbf{k}-\mathbf{k}',t')$$

where $\tilde{c}_0(\mathbf{k}, t)$ fulfils the 'unperturbed' problem, i.e. for $\tilde{\mathbf{w}}(\mathbf{k}) = \mathbf{0}$. It is the solution of the zeroth order: $\tilde{c}_0(\mathbf{k},t) = \Theta(t)\exp(-(D_0\mathbf{k}^2 - \mathrm{i}\mathbf{u}_0\cdot\mathbf{k})t) =: \Theta(t)g_0(\mathbf{k},t)$. $\Theta(t)$ denotes the Heaviside step function. Using the explicit expression for $\tilde{\mathbf{w}}$, we obtain

$$\int_{\mathbf{k}}\mathrm{i}\mathbf{k}\cdot\tilde{\mathbf{w}}(\mathbf{k}')\tilde{c}(\mathbf{k}-\mathbf{k}',t') = u_0\sqrt{\frac{q_0}{2N}}\sum_{j=1}^{N}\mathrm{i}\mathbf{k}\cdot\mathbf{p}(\mathbf{q}^{(j)})\left(\mathrm{e}^{\mathrm{i}\alpha^{(j)}}\tilde{c}(\mathbf{k}+\mathbf{q}^{(j)},t') + \mathrm{e}^{-\mathrm{i}\alpha^{(j)}}\tilde{c}(\mathbf{k}-\mathbf{q}^{(j)},t')\right)$$

and the concentration $\tilde{c}(\mathbf{k}, t)$ fulfils for $t \geqslant 0$:

$$\begin{aligned}\tilde{c}(\mathbf{k},t) = g_0(\mathbf{k},t) - \int_0^t \mathrm{d}t' g_0(\mathbf{k},t-t')\sum_{j=1}^{N}\{&A_j(\mathbf{k})\tilde{c}(\mathbf{k}+\mathbf{q}^{(j)},t') \\ &+ A_j^*(-\mathbf{k})\tilde{c}(\mathbf{k}-\mathbf{q}^{(j)},t')\}\end{aligned} \tag{12}$$

where the function $A_j(\mathbf{k})$ is defined by $A_j(\mathbf{k}) = u_0\sqrt{\frac{q_0}{2N}}\mathrm{i}\mathbf{k}\cdot\mathbf{p}(\mathbf{q}^{(j)})\,\mathrm{e}^{\mathrm{i}\alpha^{(j)}}$. Further, for $j = 1,\ldots,N$ let $\mathbf{q}^{(-j)} := -\mathbf{q}^{(j)}$ and $\alpha^{(-j)} := -\alpha^{(j)}$. Hence, it is $A_{-j}(\mathbf{k}) = A_j^*(-\mathbf{k})$, with the complex conjugate denoted by $*$, and we can deduce by (12)

$$\tilde{c}(\mathbf{k},t) = g_0(\mathbf{k},t) - \int_0^t \mathrm{d}t' g_0(\mathbf{k},t-t')\sum_{j}{}'A_j(\mathbf{k})\tilde{c}(\mathbf{k}+\mathbf{q}^{(j)},t')$$

where $\sum_j{}' := \sum_{j=-N\cdots N}$ with $j \neq 0$. Defining the function $\gamma(\mathbf{k}, t)$ which fulfils $\tilde{c}(\mathbf{k},t) =: g_0(\mathbf{k},t)\gamma(\mathbf{k},t)$, and $\sigma_j(\mathbf{k}) = D_0\mathbf{q}^{(j)^2} - \mathrm{i}\mathbf{u}_0\cdot\mathbf{q}^{(j)} + 2D_0\mathbf{k}\cdot\mathbf{q}^{(j)}$, we conclude the equation

$$\gamma(\mathbf{k},t) = 1 - \int_0^t \mathrm{d}t'\sum_{j}{}'A_j(\mathbf{k})\,\mathrm{e}^{-\sigma_j(\mathbf{k})t'}\gamma(\mathbf{k}+\mathbf{q}^{(j)},t').$$

The iteration of this equation yields a series expansion for $\gamma(\mathbf{k},t)$ in $A_j(\mathbf{k})$ or $\sqrt{q_0}$:

$$\gamma(\mathbf{k},t) = 1 - \int_0^t \mathrm{d}t' {\sum_{j_1}}' A_{j_1}(\mathbf{k})\,\mathrm{e}^{-\sigma_{j_1}(\mathbf{k})t'} + \int_0^t \mathrm{d}t' {\sum_{j_1}}' A_{j_1}(\mathbf{k})\,\mathrm{e}^{-\sigma_{j_1}(\mathbf{k})t'} \int_0^{t'} \mathrm{d}t'' {\sum_{j_2}}' A_{j_2}(\mathbf{k}+\mathbf{q}^{(j_1)})\,\mathrm{e}^{-\sigma_{j_2}(\mathbf{k}+\mathbf{q}^{(j_1)})t''} - \cdots .$$

We truncate this series after the second order in $A_j(\mathbf{k})$ and use the expression for $\gamma(\mathbf{k},t)$ to obtain the transport parameters according to equation (11):

$$u_i(t) = u_{0i} - \mathrm{i}\partial_{k_i} F(\mathbf{k},t)|_{\mathbf{k}=\mathbf{0}} \qquad D_{ij}(t) = D_0\delta_{ij} - \tfrac{1}{2}\partial_{k_i}\partial_{k_j} F(\mathbf{k},t)\big|_{\mathbf{k}=\mathbf{0}} \tag{13}$$

where $F(\mathbf{k},t) = \partial_t \ln\gamma(\mathbf{k},t)$ denotes the generating function. It is given at second order by

$$F(\mathbf{k},t) = -{\sum_{j_1}}' A_{j_1}(\mathbf{k})\,\mathrm{e}^{-\sigma_{j_1}(\mathbf{k})t} - {\sum_{j_1}}' A_{j_1}(\mathbf{k})\,\mathrm{e}^{-\sigma_{j_1}(\mathbf{k})t} \int_0^t \mathrm{d}t' {\sum_{j_2}}' A_{j_2}(\mathbf{k})\,\mathrm{e}^{-\sigma_{j_2}(\mathbf{k})t'} + {\sum_{j_1}}' A_{j_1}(\mathbf{k})\,\mathrm{e}^{-\sigma_{j_1}(\mathbf{k})t} \int_0^t \mathrm{d}t' {\sum_{j_2}}' A_{j_2}(\mathbf{k}+\mathbf{q}^{(j_1)})\,\mathrm{e}^{-\sigma_{j_2}(\mathbf{k}+\mathbf{q}^{(j_1)})t'} .$$

To calculate the transport parameters it is useful to integrate over time t and expand $F(\mathbf{k},t)$ in the components of $\mathbf{k}$ up to order $O(\mathbf{k}^2)$. Higher-order terms vanish in $u_i(t)$ and $D_{ij}(t)$.

Here and in the next sections, we use the following definitions:

$$\tau_u^{(j)} := (\mathbf{u}_0\cdot\mathbf{q}^{(j)})^{-1} = \left(u_0 q_1^{(j)}\right)^{-1} \qquad \tau_D^{(j)} := \left(D_0 \mathbf{q}^{(j)2}\right)^{-1} \qquad \tau_{D,\max} := \max_j\left(\tau_D^{(j)}\right) \tag{14}$$

for $j = \pm 1,\ldots,\pm N$. $\tau_{D,\max}$ denotes the time in which the concentration samples over the largest length period in the periodic medium in all directions due to the spreading by the local diffusion.

3.2. Centre-of-mass velocity

According to equation (13) we obtain for the centre-of-mass velocity

$$u_i(t) = u_0\delta_{1i} - u_0\sqrt{\frac{q_0}{2N}}{\sum_{j_1}}' p_i(\mathbf{q}^{(j_1)})\,\mathrm{e}^{\mathrm{i}\alpha^{(j_1)}}\,\mathrm{e}^{-\sigma_{j_1}(0)t} + u_0\sqrt{\frac{q_0}{2N}}{\sum_{j_1}}'{\sum_{j_2}}'' p_i(\mathbf{q}^{(j_1)})A_{j_2}(\mathbf{q}^{(j_1)})\,\mathrm{e}^{\mathrm{i}\alpha^{(j_1)}}\frac{\mathrm{e}^{-\sigma_{j_1}(0)t}}{\sigma_{j_2}(\mathbf{q}^{(j_1)})}\left(1-\mathrm{e}^{-\sigma_{j_2}(\mathbf{q}^{(j_1)})t}\right) \tag{15}$$

where $\sum_j'' := \sum_j'$ for $\mathbf{q}^{(j)} \neq \mathbf{0}$. It is obvious that for $t\to\infty$ the centre-of-mass velocity fulfils $\lim_{t\to\infty}\mathbf{u}(t) = \mathbf{u}_0$, and $\mathbf{u}(t)$ approaches $\mathbf{u}_0$ for $t > \tau_{D,\max}$. The ensemble mean for the randomly periodic media is given by equation (7) as $\overline{\mathbf{u}(t)} = \mathbf{u}_0$ in order $O(q_0)$. This result agrees with the result for the centre-of-mass velocity for the ensemble of the Gaussian random field, see [8]. The limit $N\to\infty$ must not be performed for $\overline{\mathbf{u}(t)}$ since all contributions do not depend on N in $O(q_0)$.

3.3. Dispersion coefficient

The detailed expression for the dispersion coefficient $D_{ij}(t)$ given by (13) can be found in appendix A.1. For $D_0 = 0$, one finds $D_{ij}(t) = 0$, as the initial point source remains point-like all the time. For $D_0 \neq 0$, we obtain $D_{ij}(t=0) = D_0\delta_{ij}$, and in the limit $t\to\infty$:

$$D_{ij}^{\infty} := D_{ij}(t\to\infty) = D_0\delta_{ij} + \frac{q_0 u_0^2}{N}{\sum_n}^* p_i(\mathbf{q}^{(n)})p_j(\mathbf{q}^{(n)})\frac{1/\tau_D^{(n)}}{\left(\tau_D^{(n)}\right)^{-2}+\left(\tau_u^{(n)}\right)^{-2}} \tag{16}$$

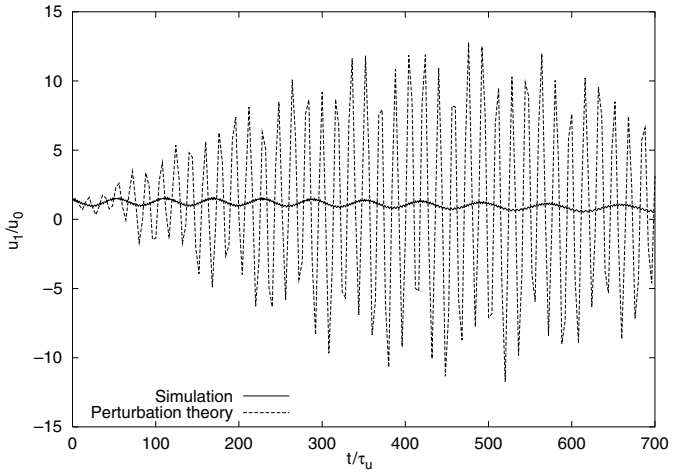


Figure 1. Comparison between $u_1(t)$ from the perturbation theory and the result from the numerical simulation for the case $D_0 = 0$ and a single realization with $N = 3$, $q_0 = 0.1$.

where $\sum_n^* := \sum_{n=1}^N$ for $\mathbf{q}^{(n)} \neq \mathbf{0}$. In the following, we concentrate on the longitudinal dispersion coefficient $D_{11}(t)$. The heterogeneity of the media determines the temporal behaviour of this coefficient in a characteristic form, whereas the contributions for the transversal dispersion coefficients are of minor importance in the approach to the order $O(q_0)$, see e.g. [8].

The ensemble mean of the longitudinal dispersion coefficient $D_{11}(t)$ agrees with the result for the ensemble of the Gaussian random field to the order $O(q_0)$, see appendix A.2 for $\overline{D_{11}(t)}$.

3.4. Comparison with numerical simulations

We consider the case $D_0 = 0$ where the transport is governed by the advection. We compare $u_1(t)$ of (15) with the result from the numerical simulation as described in section 2.3. It turns out that the perturbation theory result fails to predict the numerical simulation. It exhibits oscillations which are much larger than the numerical result as illustrated for a single realization in figure 1.

Taking the result from the iteration of the Langevin equation, see next section, the result for the velocity (15) can be related to the corresponding result $\mathbf{u}^{(2)}(t)$ from (20) if it is expanded in a series in w_i. It yields in the order $O(q_0)$

$$u_0\delta_{1i} - w_i(\mathbf{u}_0 t) + \nabla w_i(\mathbf{x})|_{\mathbf{x}=\mathbf{u}_0 t} \cdot \int_0^t \mathrm{d}t' \mathbf{w}(\mathbf{u}_0 t')$$

which is identical with the result (15) for $D_0 = 0$ using $\mathbf{w}(\mathbf{x})$. However, the expansion of $w_i(\mathbf{u}_0 t - \int_0^t \mathrm{d}t' \mathbf{w}(\mathbf{u}_0 t'))$ in (20) to the given order is a good approximation provided that w_i does not fluctuate to very near the expansion point. Due to the high fluctuations in the flow field this condition does not hold in general and the result from the perturbative expansion fails. It reveals that in the case $D_0 = 0$ the discrepancy between theory and simulation is caused by the terms of order $O(q_0)$. As can be seen in figure 1 the error is immense even for small N where the flow fluctuations are not too strong. For the averaged quantity $\overline{u(t)}$, however, the terms of $O(q_0)$ do not contribute. As a result the poor approximation by the perturbation theory is hidden comparing the ensemble mean with numerical simulations, see e.g. [11, 7].

In the case of non-zero diffusion, the agreement between $u_1(t)$ and the centre-of-mass velocity from the simulation is slightly better. This is due to the diffusion effect which acts like an error damping. However, a systematical error is still inherent in $\mathbf{u}(t)$ from the perturbation theory for $t < \tau_{D,\max}$. Analogously, the result for $D_{11}(t)$ from the perturbation analysis fails to predict the dispersion coefficient from the numerical simulation for times $t < \tau_{D,\max}$, see [11].

4. Iteration of the Langevin equation

In this section we formulate a new approach for calculating the transport parameters. We make use of the equivalence of the advection–dispersion equation and the description of the transport problem by the Langevin equation. We develop by simple iteration approximations for the centre-of-mass velocity and the dispersion coefficient for the realizations of the periodic ensemble. The theoretical approximations are again compared with the quantities of the numerical simulations.

4.1. Iterative solution of the Langevin equation

The Langevin equation for the flow field $\mathbf{u}(\mathbf{x}) = \mathbf{u}_0 - \mathbf{w}(\mathbf{x})$ is equivalent to

$$\mathbf{x}(t) = \mathbf{u}_0 t - \int_0^t \mathrm{d}t' \mathbf{w}(\mathbf{x}(t')) + \mathbf{X}_w(t) \tag{17}$$

where $\mathbf{x}(t = 0) = \mathbf{0}$ und $\mathbf{X}_w(t) := \int_0^t \mathrm{d}t' \boldsymbol{\xi}(t')$ for a realization of the white noise $\boldsymbol{\xi}(t)$. In the case of a homogeneous velocity field, i.e. $\mathbf{w}(\mathbf{x}) = \mathbf{0}$, the solution is given by $\mathbf{x}^{(0)}(t) = \mathbf{u}_0 t + \mathbf{X}_w(t)$. An iteration of equation (17) yields an approximation for the solution of the Langevin equation, whereby the solution $\mathbf{x}^{(0)}(t)$ is substituted as an approximation for $\mathbf{x}(t')$. Thus, the approximative trajectory $\mathbf{x}(t)$ is given due to one iteration step by

$$\mathbf{x}^{(1)}(t) = \mathbf{u}_0 t + \mathbf{X}_w(t) - \int_0^t \mathrm{d}t' \mathbf{w}(\mathbf{u}_0 t' + \mathbf{X}_w(t')). \tag{18}$$

Analogously, a two-fold iteration of (17) yields

$$\mathbf{x}^{(2)}(t) = \mathbf{u}_0 t + \mathbf{X}_w(t) - \int_0^t \mathrm{d}t' \mathbf{w}\left(\mathbf{u}_0 t' + \mathbf{X}_w(t') - \int_0^{t'} \mathrm{d}t'' \mathbf{w}(\mathbf{u}_0 t'' + \mathbf{X}_w(t''))\right). \tag{19}$$

$\mathbf{x}^{(1)}(t)$ and $\mathbf{x}^{(2)}(t)$ can be understood as an approximation of the solution of the Langevin equation (8) for a single realization of the white noise. They are used to obtain the transport parameters for a realization of the ensemble of periodic fields. For the case of $D_0 = 0$ the transport parameters are solely determined by the advection. We derive an approximation for this case by (19) which agrees with the numerical simulations in contrast to the result from the perturbation theory. Similar approximations are then derived for the case $D_0 \neq 0$.

4.2. Parameters for vanishing diffusion

For vanishing diffusion, the terms of the white noise vanish in the expressions for $\mathbf{x}^{(1)}(t)$ and $\mathbf{x}^{(2)}(t)$, i.e. $\mathbf{X}_w(t) = \mathbf{0}$. We denote the trajectory for $D_0 = 0$ by $\langle \mathbf{x}(t) \rangle_0$. Using (18) respectively (19) the trajectory is given by

$$\langle \mathbf{x}^{(1)}(t) \rangle_0 = \mathbf{u}_0 t - \int_0^t \mathrm{d}t' \mathbf{w}(\mathbf{u}_0 t') \qquad \langle \mathbf{x}^{(2)}(t) \rangle_0 = \mathbf{u}_0 t - \int_0^t \mathrm{d}t' \mathbf{w}\left(\mathbf{u}_0 t' - \int_0^{t'} \mathrm{d}t'' \mathbf{w}(\mathbf{u}_0 t'')\right).$$

We substitute the fluctuations $\mathbf{w}(\mathbf{x})$ from equation (5) and perform the time integration to obtain

$$\langle \mathbf{x}^{(1)}(t)\rangle_0 = \mathbf{u}_0 t - u_0\sqrt{\frac{2q_0}{N}}\sum_{j=1}^{N}\mathbf{p}(\mathbf{q}^{(j)})(\mathbf{q}^{(j)}\cdot\mathbf{u}_0)^{-1}(\sin(\mathbf{q}^{(j)}\mathbf{u}_0 t+\alpha^{(j)})-\sin\alpha^{(j)}).$$

Analogously, the approximation $\langle \mathbf{x}^{(2)}(t)\rangle_0$ is given by $\langle \mathbf{x}^{(2)}(t)\rangle_0 = \mathbf{u}_0 t - \int_0^t \mathrm{d}t'\mathbf{w}(\langle \mathbf{x}^{(1)}(t')\rangle_0)$, where the time integration cannot be performed analytically. Differentiating the given approximations with respect to time, we obtain the centre-of-mass velocity. As the point injection does not spread for $D_0 = 0$ the centre-of-mass velocity is the velocity of the injection. The first iteration $\langle \mathbf{x}^{(1)}(t)\rangle_0$ yields

$$\mathbf{u}^{(1)}(t) = \mathbf{u}_0 - u_0\sqrt{\frac{2q_0}{N}}\sum_{j=1}^{N}\mathbf{p}(\mathbf{q}^{(j)})\cos\left(t\big/\tau_u^{(j)}+\alpha^{(j)}\right)$$

with time scales $\tau_u^{(j)}$ given by (14). $\mathbf{u}^{(1)}(t)$ is the flow velocity at the location $\mathbf{x} = \mathbf{u}_0 t$, which is a simple approximation for the centre-of-mass velocity due to the fluctuations of the flow field. The second iteration of the Langevin equation yields for the centre-of-mass velocity

$$\mathbf{u}^{(2)}(t) = \mathbf{u}_0 - \mathbf{w}\left(\mathbf{u}_0 t - u_0\sqrt{\frac{2q_0}{N}}\sum_{j=1}^{N}\mathbf{p}(\mathbf{q}^{(j)})\tau_u^{(j)}\left(\sin\left(t\big/\tau_u^{(j)}+\alpha^{(j)}\right)-\sin\alpha^{(j)}\right)\right). \tag{20}$$

The result $\mathbf{u}^{(2)}(t)$ approximates the velocity better than $\mathbf{u}^{(1)}(t)$ as it incorporates the impact of the flow fluctuations on the trajectory of the particle. Figure 2 depicts the quantities $u_1^{(1)}(t)$ and $u_1^{(2)}(t)$ for a single realization in comparison with the result of the numerical simulation where we consider only the component of the velocity in the direction of the drift (1-direction). $u_1^{(1)}(t)$ compared to the result from the simulation shows that the initial agreement between both quantities becomes less for growing time owing to the simple approximation of the particles' location. Thereas, $u_1^{(2)}(t)$ indicates a good agreement for large times, see figure 2(*b*).

The time-averaged square deviation between the approximation $u_{\mathrm{app}}(t)$ and the simulation $u_{\mathrm{sim}}(t)$ for M time steps $\eta := (M\Delta t)^{-1}\sum_{m=1}^{M}\sqrt{(u_{\mathrm{app}}(t_m)-u_{\mathrm{sim}}(t_m))^2}$ and $t_m = m\Delta t$, yields for $u_1^{(1)}(t)$ and $u_1^{(2)}(t)$: $\eta^{(1)} = 0.187u_0$ and $\eta^{(2)} = 0.117u_0$ ($\Delta t = \tau_u$, $M = 200$).

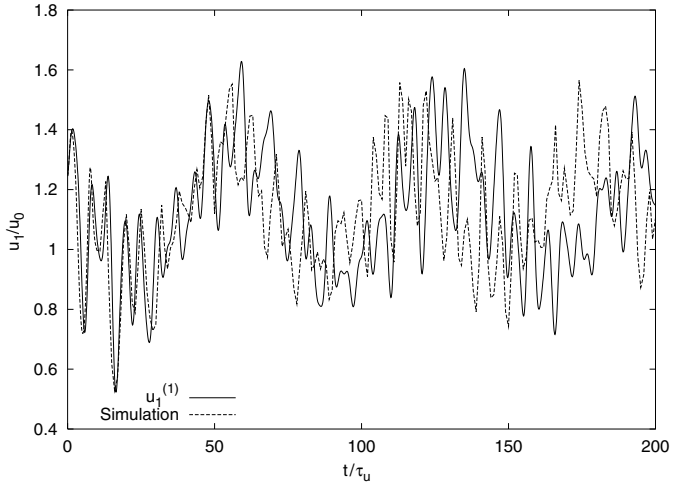
We conclude that in the case $D_0 = 0$ and small q_0 iterating the Langevin equation yields good approximations for the centre-of-mass velocity in contrast to the perturbation theory analysis.

The analogous expression for the dispersion coefficient is due to (1) given as $D_{ij}(t) \equiv 0$ for $D_0 = 0$. So, the point injection remains point-like for all times t.

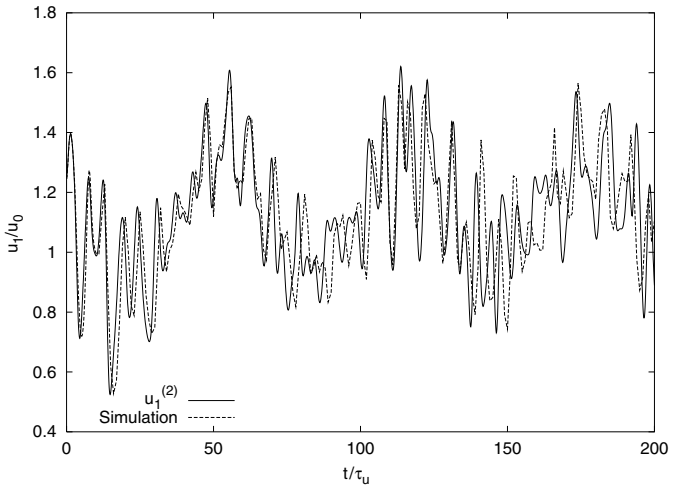
4.3. Parameters for $D_0 \neq 0$

To derive approximations in the case $D_0 \neq 0$ we apply only the first iterative solution of the Langevin equation. In this case, the ensemble average for the white noise can be performed analytically. The approximation for a realization of the noise reads

$$\mathbf{x}^{(1)}(t) = \mathbf{u}_0 t + \mathbf{X}_w(t) - \int_0^t \mathrm{d}t'\int_{\mathbf{k}} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot(\mathbf{u}_0 t'+\mathbf{X}_w(t'))}\tilde{\mathbf{w}}(\mathbf{k}). \tag{21}$$



(*a*)



(*b*)

Figure 2. $u_1^{(1)}(t)$ (*a*) and $u_1^{(2)}(t)$ (*b*) in comparison with $u_1(t)$ from the numerical simulation for a single realization with $N = 10$ and $q_0 = 0.1$.

4.3.1. Centre-of-mass velocity. According to (21) the noise averaged trajectory is given by

$$\langle \mathbf{x}^{(1)}(t)\rangle = \mathbf{u}_0 t - \int_0^t \mathrm{d}t' \int_{\mathbf{k}} \tilde{\mathbf{w}}(\mathbf{k})\, \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{u}_0 t'} \left\langle \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\int_0^{t'} \mathrm{d}t''\xi(t'')}\right\rangle$$

as $\langle \mathbf{X}_w(t)\rangle = \mathbf{0}$. The expression $\left\langle \mathrm{e}^{-\mathrm{i}\mathbf{k}\int_0^{t'} \mathrm{d}t''\xi(t'')}\right\rangle$ is given by a functional integral according to (9). As shown in appendix C, the following equality holds for $D_{ij} = D_0\delta_{ij}$: $\left\langle \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\int_0^{t} \mathrm{d}t'\xi(t')}\right\rangle = \exp(-D_0\mathbf{k}^2 t)$. This leads to

$$\langle \mathbf{x}^{(1)}(t)\rangle = \mathbf{u}_0 t - \int_0^t \mathrm{d}t' \int_{\mathbf{k}} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{u}_0 t'}\, \mathrm{e}^{-D_0\mathbf{k}^2 t'} \tilde{\mathbf{w}}(\mathbf{k}).$$

The $\mathbf{k}$-integration and the time integration can be performed using the explicit expression for $\tilde{\mathbf{w}}(\mathbf{k})$ of equation (6), and the centre-of-mass velocity in a single realization of the periodic

ensemble is given by

$$\mathbf{u}(t) = \mathbf{u}_0 - u_0\sqrt{\frac{2q_0}{N}}\sum_{j=1}^{N}\mathbf{p}(\mathbf{q}^{(j)})\,\mathrm{e}^{-t/\tau_D^{(j)}}\cos\left(t\big/\tau_u^{(j)} + \alpha^{(j)}\right)$$

which yields $\lim_{t\to\infty}\mathbf{u}(t) = \mathbf{u}_0$. The ensemble average for the periodic fields also produces $\overline{\mathbf{u}(t)} = \mathbf{u}_0$. Therefore, $\mathbf{u}(t)$ tends to $\mathbf{u}_0$ for $t \gg \tau_{D,\max}$ because of the enlargement of the plume by diffusion which results in the limit $t \to \infty$ in an equivalence of volume and ensemble averaging. The result given by the ensemble mean agrees with the result for the ensemble of a Gaussian random field, see e.g. [8]. The transition $N \to \infty$ must not be performed for the same reason as for the result of the perturbation theory. Furthermore, the result for $\overline{\mathbf{u}(t)}$ and $\mathbf{u}(t \to \infty)$ is valid for all realizations of the ensemble of periodic media since it is independent of $\{\mathbf{q}^{(j)}, \alpha^{(j)}, N\}$.

In figure 3(*a*) the centre-of-mass velocity $u_1(t)$ is plotted in comparison to the result of the numerical simulation for one realization of the periodic ensemble. It shows that both quantities are in good agreement for times $t \ll \tau_{D,\max}$ and $t > \tau_{D,\max}$. This can be understood since the approximation for small t is given by the result for $D_0 = 0$. Further, for large time scales the asymptotic value is approached by $u_1(t)$. For times between these regimes, the approximative result is not the best since the first iterative solution of the Langevin equation does not account for the flow fluctuations. To account for them one would have to consider the second iterative solution. Unfortunately, for the latter the integrations for the white noise cannot be performed analytically.

4.3.2. Dispersion coefficient. Analogously, we get an approximation for the dispersion coefficient by the first iterative solution. Its longitudinal component results (with the aid of $\mathbf{x}^{(1)}(t)$, equation (21), and the definition for $D_{11}(t)$) in

$$D_{11}(t) = D_0 + \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\left\langle\left(\int_0^t \mathrm{d}t' w_1(\mathbf{u}_0 t' + \mathbf{X}_w(t'))\right)^2\right\rangle - \frac{\mathrm{d}}{\mathrm{d}t}\left\langle X_{w1}(t)\int_0^t \mathrm{d}t' w_1(\mathbf{u}_0 t' + \mathbf{X}_w(t'))\right\rangle - \frac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}\left(\int_0^t \mathrm{d}t'\langle w_1(\mathbf{u}_0 t' + \mathbf{X}_w(t'))\rangle\right)^2.$$

In order to perform the average over the ensemble of the noise analytically, we make use of the Fourier transform for $w_1(\mathbf{x})$. Using $\mathbf{X}_w(t) = \int_0^t \mathrm{d}t'\boldsymbol{\xi}(t')$, we obtain

$$\begin{aligned} D_{11}(t) = D_0 &+ \int_0^t \mathrm{d}t' \int_{\mathbf{k}}\int_{\mathbf{k}'} \tilde{w}_1(\mathbf{k})\tilde{w}_1(\mathbf{k}')\,\mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t+\mathbf{k}'t')}\left\langle \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\int_0^t \mathrm{d}t''\boldsymbol{\xi}(t'') - \mathrm{i}\mathbf{k}'\cdot\int_0^{t'}\mathrm{d}t''\boldsymbol{\xi}(t'')}\right\rangle \\ &- \frac{\mathrm{d}}{\mathrm{d}t}\int_0^t \mathrm{d}t'\int_0^t \mathrm{d}t''\int_{\mathbf{k}}\tilde{w}_1(\mathbf{k})\,\mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot\mathbf{k}t'}\left\langle \xi_1(t'')\,\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\int_0^{t'}\mathrm{d}t'''\boldsymbol{\xi}(t''')}\right\rangle \\ &- \int_0^t \mathrm{d}t' \int_{\mathbf{k}}\int_{\mathbf{k}'} \tilde{w}_1(\mathbf{k})\tilde{w}_1(\mathbf{k}')\,\mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t+\mathbf{k}'t')}\left\langle \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\int_0^t \mathrm{d}t''\boldsymbol{\xi}(t'')}\right\rangle\left\langle \mathrm{e}^{-\mathrm{i}\mathbf{k}'\cdot\int_0^{t'}\mathrm{d}t''\boldsymbol{\xi}(t'')}\right\rangle. \end{aligned} \tag{22}$$

The integrations in $D_{11}(t)$ can be performed, as shown in appendix C, and is as follows:

$$\begin{aligned} D_{11}(t) = D_0 &+ \int_0^t \mathrm{d}t' \int_{\mathbf{k}}\int_{\mathbf{k}'} \tilde{w}_1(\mathbf{k})\tilde{w}_1(\mathbf{k}')\,\mathrm{e}^{-D_0(\mathbf{k}^2 t + \mathbf{k}'^2 t' + 2\mathbf{k}\cdot\mathbf{k}'t')}\,\mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t+\mathbf{k}'t')} \\ &+ 2\mathrm{i}D_0 t\int_{\mathbf{k}}\tilde{w}_1(\mathbf{k})k_1\,\mathrm{e}^{-D_0\mathbf{k}^2 t - \mathrm{i}\mathbf{u}_0\cdot\mathbf{k}t} \\ &- \int_0^t \mathrm{d}t' \int_{\mathbf{k}}\int_{\mathbf{k}'} \tilde{w}_1(\mathbf{k})\tilde{w}_1(\mathbf{k}')\,\mathrm{e}^{-D_0(\mathbf{k}^2 t + \mathbf{k}'^2 t')}\,\mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t+\mathbf{k}'t')}. \end{aligned}$$

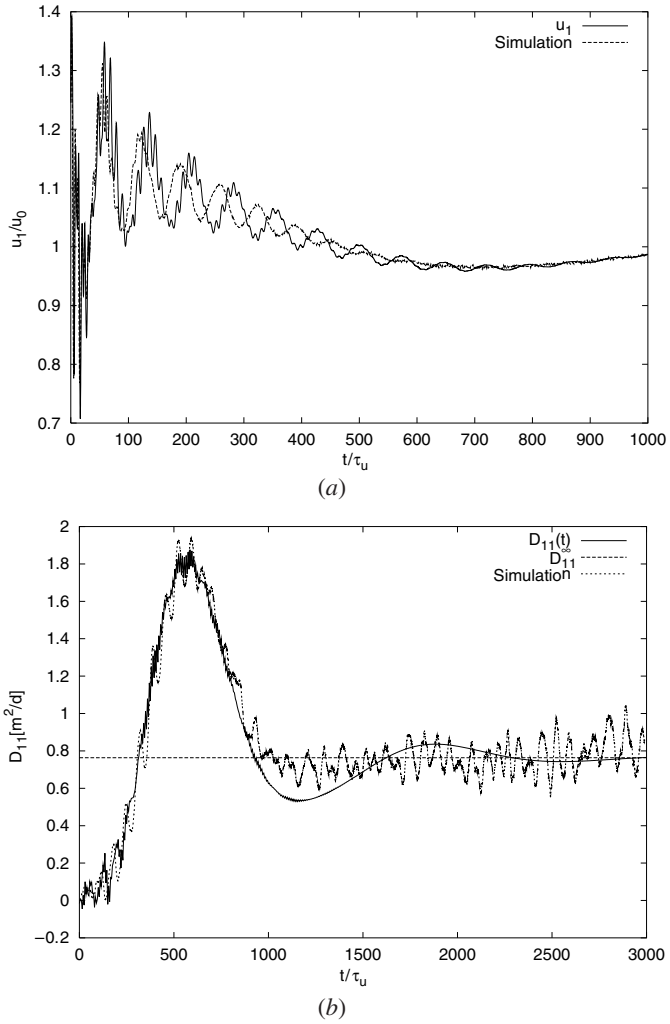


Figure 3. Transport parameters from the iterative solution of the Langevin equation: (*a*) $u_1(t)$ and the numerical result for the centre-of-mass velocity for one realization and (*b*) $D_{11}(t)$ and D_{11}^{∞} compared to the smoothed result for $D_{11}(t)$ from the numerical simulation. The parameters of the single realization are: $N = 10, q_0 = 0.1, D_0 = 0.01m^2/d, \tau_{D,\max}/\tau_u = 552$, and for the numerical simulation: $R = 10\,000$.

The $\mathbf{k}$- and $\mathbf{k}'$-integrations can be performed using $\tilde{w}_1(\mathbf{k})$ of equation (6), and the time integral can be also calculated. The full expression for $D_{11}(t)$ can be found in appendix B.1.

For $D_0 = 0$, $D_{11}(t)$ vanishes, and for $t \to \infty$ the expression of the perturbation theory for D_{11}^{∞} holds, see equation (16). $D_{11}(t)$ approaches D_{11}^{∞} for time scales $t \gg \tau_{D,\max}$. So, the behaviour of the dispersion coefficient is determined for finite times as well as in the limit $t \to \infty$ by the parameters $\{\mathbf{q}^{(j)}, \alpha^{(j)}, N\}$. For $N < \infty$ $D_{11}(t)$ depends on the structure of the realization of the flow field. Nevertheless, the ensemble average for $D_{11}(t)$ in the order $O(q_0)$ yields the same result $\overline{D_{11}(t)}$ as for the ensemble of the Gaussian random field, see appendix B.2 and [8]. Again, the transition $N \to \infty$ can be neglected since $\overline{D_{11}(t)}$ is independent of N.

The comparison of $D_{11}(t)$ and D_{11}^{∞} given by the iterative solution of the Langevin equation with the result of numerical simulations shows that they agree well. Due to the finite number R of realizations of the noise, the simulated quantities are noisy for $t > \tau_{D,\max}$, and we apply the moving averaging for smoothing, as described in section 2. Figure 3(*b*) displays $D_{11}(t)$ for a single realization and the corresponding quantity of the numerical simulation. Unlike the result of the perturbation theory, $D_{11}(t)$ agrees with the simulation for times $t < \tau_{D,\max}$.

In the following we only consider the approximations given by the iteration of the Langevin equation for the transport parameters.

5. Self-averaging property of the velocity

We investigate the self-averaging property of the centre-of-mass velocity, that is, to what extent the ensemble average is representative of the velocity found in a single realization. Therefore, we derive the temporal behaviour of the sample-to-sample fluctuations of $u_i(t)$ by the approximation from the Langevin equation, and compare it with numerical simulations. The asymptotic value is given by $\lim_{t\to\infty} \mathbf{u}(t) = \mathbf{u}_0$. It does not depend on the number of modes nor on the given realization and is identical to the ensemble mean. This indicates that $\mathbf{u}(t)$ is self-averaging in the limit $t \to \infty$. It holds for the ensemble of randomly periodic media as well as for the ensemble of the Gaussian random field since the results do not depend on N.

The sample-to-sample fluctuations of $u_1(t)$ are given by

$$(\delta u_1(t))^2 := \overline{(u_1(t) - \overline{u_1(t)})^2} = \frac{2q_0u_0^2}{N(2\pi)^N} \int_0^{2\pi} \mathrm{d}\alpha^{(1)} \cdots \int_0^{2\pi} \mathrm{d}\alpha^{(N)} \frac{l_0^{3N}}{(2\pi)^{\frac{3N}{2}}} \int \mathrm{d}^3q^{(1)} \cdots$$
$$\times \int \mathrm{d}^3q^{(N)}\, \mathrm{e}^{-\frac{l_0^2}{2}(\mathbf{q}^{(1)^2}+\cdots+\mathbf{q}^{(N)^2})} \left(\sum_{j=1}^{N} p_1(\mathbf{q}^{(j)})\, \mathrm{e}^{-D_0\mathbf{q}^{(j)^2}t} \cos\left(u_0 q_1^{(j)} t + \alpha^{(j)}\right) \right)^2 .$$

Performing the integrations for the phases $\alpha^{(j)}$ yields

$$(\delta u_1(t))^2 = \frac{q_0u_0^2l_0^{3N}}{N(2\pi)^{\frac{3N}{2}}} \int \mathrm{d}^3q^{(1)} \cdots \int \mathrm{d}^3q^{(N)}\, \mathrm{e}^{-\frac{l_0^2}{2}\left(\mathbf{q}^{(1)^2}+\cdots+\mathbf{q}^{(N)^2}\right)} \sum_{j=1}^{N} p_1^2(\mathbf{q}^{(j)})\, \mathrm{e}^{-2D_0\mathbf{q}^{(j)^2}t}$$
$$= q_0u_0^2 \frac{l_0^3}{(2\pi)^{\frac{3}{2}}} \int \mathrm{d}^3q\, \mathrm{e}^{-\frac{l_0^2}{2}\mathbf{q}^2}\, \mathrm{e}^{-2D_0\mathbf{q}^2t}\, p_1^2(\mathbf{q})$$

which can be easily calculated. Analogously, the sample-to-sample fluctuations can be derived for the directions $i = 2, 3$, and the final result is given by

$$(\delta u_i(t))^2 = \overline{(u_i(t) - \overline{u_i(t)})^2} = \frac{q_0u_0^2S_i}{15\sqrt{1 + 4t/\tau_D}^3} \tag{23}$$

where the constant S_i is $S_i = 8$ for $i = 1$, $S_i = 1$ for $i = 2, 3$, and $\tau_D := l_0^2/D_0$ fixes the diffusive time scale.

The fluctuations $(\delta u_i(t))^2$ are independent of N and agree with the result derived by Dentz in [6] with the aid of the perturbation theory for the ensemble of a Gaussian random field. So, the self-averaging property for the centre-of-mass velocity in both cases is identical. As $(\delta u_i(t))^2$ tends monotonously to zero for increasing t and $D_0 \neq 0$, the velocity is self-averaging for time scales $t > \tau_D$. For the case $D_0 = 0$, the sample-to-sample fluctuations remain constant. This is due to the fact that the injection remains point-like and the motion is governed solely by the advection. In figure 4(*a*), $(\delta u_1(t))^2$ is plotted in comparison with

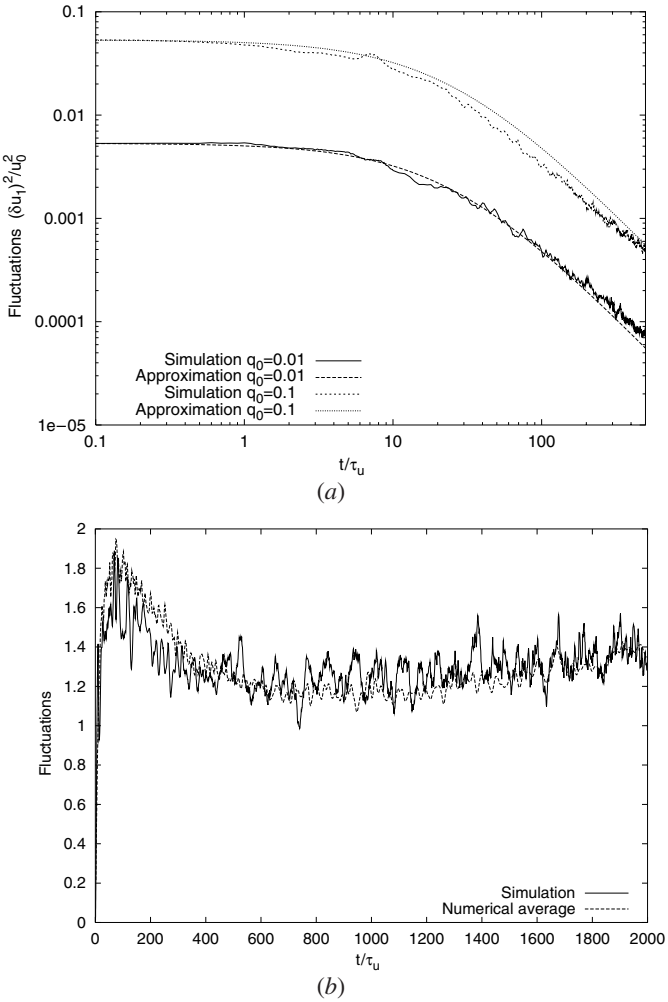


Figure 4. Comparison of the mean square sample-to-sample fluctuations by the iterative solution of the Langevin equation with the numerical simulations, $\tau_D/\tau_u = 100$. (*a*) $(\delta u_1(t))^2/\overline{u_1(t)}^2$ given by equation (23) and by the simulation for $N = 10$. For $q_0 = 0.01$ the numerical average was performed over 373 realizations and $R = 10\,000$ realizations of the white noise, and for $q_0 = 0.1$ over 1250 realizations with $R = 1000$ and (*b*) $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ given by numerical averaging of $D_{11}(t)$ over 5000 realizations and given by the simulation over 1550 realizations for $N = 50$, $q_0 = 0.1$, $R = 5000$. The results of the simulations are smoothed by a moving average.

the result from the numerical simulation for $q_0 = 0.01$ and $q_0 = 0.1$. Both quantities agree very well. As shown in [11], the result of the simulation tends even faster to zero than the theoretical value for $t > \tau_u$ and higher values of q_0.

6. Self-averaging property of the dispersion coefficient

In this section we analyse the self-averaging properties of the longitudinal dispersion coefficient given by the approximation which comes from the Langevin equation. We study the sample-to-sample fluctuations of $D_{11}(t)$ while the number of modes N tends to infinity. In the

limit $N \to \infty$ we obtain the self-averaging property of the ensemble of the Gaussian random field.

In lowest order perturbation theory the mean square sample-to-sample fluctuations of $D_{11}(t)$ were already derived in [6] for the case of the Gaussian random field:

$$\overline{(D_{11}(t) - \overline{D_{11}(t)})^2} = q_0 u_0^2 l_0^2 \frac{32}{35} \frac{(t/\tau_D)^2}{\sqrt{1 + 4t/\tau_D}^5}.$$

Compared to numerical simulations, this result underestimates the sample-to-sample fluctuations. To get a more precise result by the perturbation theory approach, one would have to take into account higher-order contribution which is very laborious. For this reason the expression for $D_{11}(t)$ given by the Langevin equation is very useful as it incorporates higher-order contributions.

In contrast to the centre-of-mass velocity, the dispersion coefficient $D_{11}(t)$ depends on the given aquifer realization for $t \to \infty$ and for finite N, see equation (16) for D_{11}^{∞}. Since $\overline{D_{11}(t)}$ is independent on N, it is clear that $D_{11}(t)$ is not self-averaging in the limit $t \to \infty$ for finite N.

6.1. Sample-to-sample fluctuations of D_{11}^{∞}

For the calculation of $(\delta D_{11}^{\infty})^2 = \overline{(D_{11}^{\infty} - \overline{D_{11}^{\infty}})^2}$ in the case of finite N, we have to regularize the integration to avoid divergent contributions in the expression for

$$(\delta D_{11}^{\infty})^2 = \int_{-\infty}^{\infty} \mathrm{d}\alpha^{(1)} P_{\alpha}(\alpha^{(1)}) \cdots \int_{-\infty}^{\infty} \mathrm{d}\alpha^{(N)} P_{\alpha}(\alpha^{(N)}) \times \int \mathrm{d}^3 q^{(1)} P_{\mathbf{q}}(\mathbf{q}^{(1)}) \cdots \int \mathrm{d}^3 q^{(N)} P_{\mathbf{q}}(\mathbf{q}^{(N)}) (D_{11}^{\infty} - \overline{D_{11}^{\infty}})^2 \tag{24}$$

for small wave vectors $\mathbf{q}$. We restrict the integration over the components $q_1^{(j)}$ by introducing a cut-off parameter L and substitute $P_{\mathbf{q}}(\mathbf{q}^{(j)})$ by a regularized distribution $\tilde{P}_{\mathbf{q}}(\mathbf{q}^{(j)})$:

$$P_{\mathbf{q}}(\mathbf{q}) \to \tilde{P}_{\mathbf{q}}(\mathbf{q}) := \mathcal{N}(L/l_0)\, \mathrm{e}^{-\frac{1}{2} l_0^2 \mathbf{q}^2} \left(1 - \Theta\left(\frac{l_0^2}{L^2} - \frac{q_1^2}{\mathbf{q}^2}\right)\right).$$

The normalization factor $\mathcal{N}(L/l_0)$ is chosen so that $\int \mathrm{d}^3 q\, \tilde{P}_{\mathbf{q}}(\mathbf{q}) = 1$. For $L \to \infty$ we have then $\tilde{P}_{\mathbf{q}}(\mathbf{q}) \to P_{\mathbf{q}}(\mathbf{q})$. Using the modified distribution $\tilde{P}_{\mathbf{q}}(\mathbf{q})$ the integral in (24) remains finite, and the result for $(\delta D_{11}^{\infty})^2$ reads

$$(\delta D_{11}^{\infty})^2 = \frac{G(\epsilon)}{N} + \frac{q_0^2 u_0^2 l_0^2}{4\epsilon N} \sqrt{\frac{\pi}{2}} \ln(L/l_0) \tag{25}$$

where the function $G(\epsilon)$ does not depend on L and is of order $O\left(q_0^2\right)$, $\epsilon := \tau_u/\tau_D = D_0/u_0 l_0$. The detailed analysis of (25) can be found in appendix B.3. Due to the contributions of order $O\left(q_0^2\right)$ in the sample-to-sample fluctuations, it is necessary to account for the four-point correlation function of the flow fluctuations $\mathbf{w}(\mathbf{x})$. The result proves that $(\delta D_{11}^{\infty})^2$ vanishes for $N \to \infty$. Thus, for an ensemble of the Gaussian random field the asymptotic, longitudinal dispersion coefficient is self-averaging. This does not hold for the ensemble of randomly periodic media where N is finite.

A similar investigation for $(\delta D_{11}(t))^2 = \overline{(D_{11}(t) - \overline{D_{11}(t)})^2}$ yields for the asymptotic behaviour for large times $t \gg \tau_D$

$$(\delta D_{11}(t))^2 \sim \frac{q_0^2 u_0^2 l_0^2}{4\epsilon N} \sqrt{\frac{\pi}{2}} \ln(t/\tau_D). \tag{26}$$

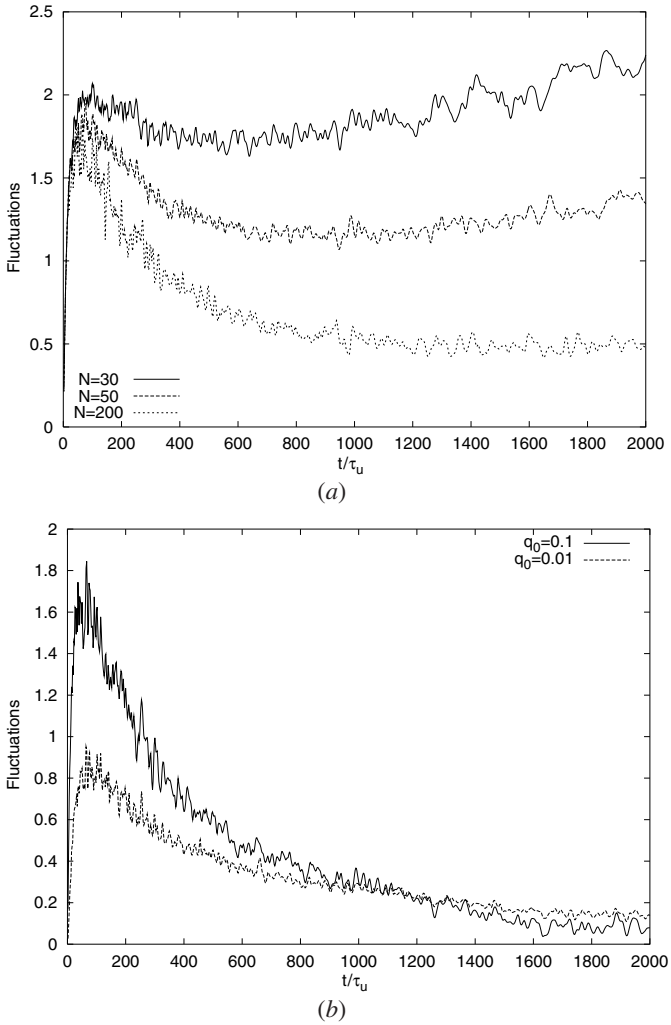


Figure 5. $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ from numerical averaging of $D_{11}(t)$ from the Langevin equation, $\tau_D/\tau_u = 100$. (*a*) For $N = 30$ with 10 000 realizations, $N = 50$ with 5000 realizations, and $N = 200$ with 1000 realizations, $q_0 = 0.1$. (*b*) Extrapolated to $N \to \infty$ from the numerically averaged sample-to-sample fluctuations of $D_{11}(t)$. The extrapolation was performed with the results for $N = 10, 20, 30, 50, 100, 200$ and $N = 1000$.

This result again depends on the number of modes and has a logarithmic divergence in time, but vanishes for infinite N. However, for the ensemble of the Gaussian random field, i.e. for $N \to \infty$, the sample-to-sample fluctuations $(\delta D_{11}(t))^2$ tend to zero in the limit $t \to \infty$ due to (25).

6.2. Computation of the sample-to-sample fluctuations

In figure 4(*b*) the fluctuations $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ from the numerical averaging of $D_{11}(t)$, equation (B1), are plotted for 5000 realizations of the ensemble of periodic fields with $N = 50$, and the result obtained from the numerical simulation, as described in section 2. In the

numerical simulation we used 1550 realizations with $R = 5000$ realizations of the white noise. Furthermore, the dispersion coefficient of each realization was smoothed for computing $(\delta D_{11}(t))^2$. Both quantities agree qualitatively well. The asymptotic increase with time due to (26) is also noticeable.

In the following we restrict ourselves to the numerical evaluation of $(\delta D_{11}(t))^2$ given by the theoretical approximation of the Langevin equation, equation (B1), and analyse the sample-to-sample fluctuations for growing N, as the direct computation by the numerical simulation for comparisons is very costly. A qualitative analysis of $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ given by the numerical average over $D_{11}(t)$ for increasing $N < \infty$ exhibits the following: The sample-to-sample fluctuations have a local minimum in the range $t \approx \tau_D$, which does not change with N. Further, the slow rise in $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ due to the asymptotic value is shifted to larger time scales for increasing N. So, for the ensemble of periodic fields the fluctuations first become smaller for $t > \tau_D$ owing to the dispersion but they grow for larger times due to (26). In this case the dispersion coefficient is not self-averaging at all. Figure 5(a) shows $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ for different values of N.

To obtain the sample-to-sample fluctuations for the Gaussian random ensemble the results are extrapolated to $N \to \infty$ using the dependence of $1/N$. Figure 5(b) shows $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2$ given by the numerical averaging which is extrapolated to $N \to \infty$. The plot includes the results for $q_0 = 0.01$ and $q_0 = 0.1$. It shows that the dispersion coefficient $D_{11}(t)$ becomes more and more self-averaging for $t > \tau_D$ for the ensemble of the Gaussian field which agrees with the above asymptotic result. However, for $q_0 = 0.1$ the fluctuations are small only for times larger than one order of magnitude of τ_D. Therefore, the predictive ability of the stochastic approach is poor for the Gaussian ensemble.

7. Conclusion

The paper investigates the behaviour of transport parameters in heterogeneous media with a flow modelled by an ensemble of periodic and Gaussian random fields. The transport parameters are constructed by appropriately defined averages over the ensembles. We calculate the centre-of-mass velocity and the dispersion coefficient of concentration distributions in given realizations using approximations based on a perturbative expansion for the transport equation, and on the iterative solution of the equivalent Langevin equation. We compare the results with numerical simulations of the transport processes. We find that the perturbation theory reproduces the numerical results only poorly, whereas the iterative solution yields good results for the behaviour in a single realization. Using the approximations obtained from the iteration we study the self-averaging properties. We find that the centre-of-mass velocity is self-averaging for $t \to \infty$. The ensemble average of the velocity characterizes the corresponding behaviour of a single realization for sufficiently large times in both kinds of ensembles. The dispersion coefficient is not self-averaging in the ensemble of periodic fields. For the Gaussian ensemble, on the other hand, the asymptotic value of the dispersion coefficient is self-averaging for $t \to \infty$. However, since the self-averaging becomes relevant first for very large times $t > 10\tau_D$ the predictive ability of the ensemble average for the dispersion coefficient in a single realization is poor for real systems. Since we discuss merely point-like injections, most experimental data cannot be evaluated to apply the new results. However, considering geostatistical data similar to those found in the Borden field site [21], that is, $q_0 \approx 0.1, u_0 \approx 0.086\,m/d$, $\tau_u = 85d$ and $\tau_D \approx 10^4 d$ which is comparable to the transversal and vertical dispersive time scales of the Borden aquifer (for a summary of the data converted for the Gaussian correlation function, see [8]), we get a self-averaging

property $(\delta D_{11}(t))^2/\overline{D_{11}(t)}^2 < 0.1$ for times $t \gtrsim 1580\tau_u$. This result clearly indicates that the predictive value of the observable is small for practical time scales in a real system.

Appendix A. Transport parameters from the perturbation theory

Appendix A.1. Dispersion coefficient

The explicit dispersion coefficient for a single realization of the ensemble of periodic media is given by

$$
\begin{aligned}
D_{11}(t) = D_0 + u_0\sqrt{\frac{2q_0}{N}}D_0 t \sum_{j_1}{}' q_1^{(j_1)} p_1(\mathbf{q}^{(j_1)})\,\mathrm{e}^{-t/\tau_D^{(j_1)}}\,\sin\left(t\big/\tau_u^{(j_1)} + \alpha^{(j_1)}\right) \\
+ \frac{u_0^2 q_0}{2N}\sum_{j_1}{}'\sum_{j_2}{}'' \frac{p_1(\mathbf{q}^{(j_1)})\,\mathrm{e}^{-t/\tau_D^{(j_1)}}}{B_{j_2}(\mathbf{q}^{(j_1)})}\Bigg\{ p_1(\mathbf{q}^{(j_2)})\big(E_{j_2}(\mathbf{q}^{(j_1)})C_{j_1,j_2}(0) - \mathbf{u}_0\cdot\mathbf{q}^{(j_2)} \\
\times S_{j_1,j_2}(0) - \mathrm{e}^{-E_{j_2}(\mathbf{q}^{(j_1)})t}\big(E_{j_2}(\mathbf{q}^{(j_1)})C_{j_1,j_2}(\mathbf{q}^{(j_2)}) - \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}S_{j_1,j_2}(\mathbf{q}^{(j_2)})\big)\big) \\
- 2D_0[\mathbf{q}^{(j_1)}\cdot\mathbf{p}(\mathbf{q}^{(j_2)})]\Bigg[\left(q_1^{(j_2)}\frac{E_{j_2}(\mathbf{q}^{(j_1)})}{B_{j_2}(\mathbf{q}^{(j_1)})} + q_1^{(j_1)}t\right)\big(E_{j_2}(\mathbf{q}^{(j_1)})C_{j_1,j_2}(0) \\
- \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}S_{j_1,j_2}(0)\big) - q_1^{(j_2)}\frac{\mathbf{u}_0\cdot\mathbf{q}^{(j_2)}}{B_{j_2}(\mathbf{q}^{(j_1)})}\big(E_{j_2}(\mathbf{q}^{(j_1)})S_{j_1,j_2}(0) + \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}C_{j_1,j_2}(0)\big) \\
- \mathrm{e}^{-E_{j_2}(\mathbf{q}^{(j_1)})t}\Bigg(\left(q_1^{(j_2)}\frac{E_{j_2}(\mathbf{q}^{(j_1)})}{B_{j_2}(\mathbf{q}^{(j_1)})} + q_1^{(j_1)}t\right)\big(E_{j_2}(\mathbf{q}^{(j_1)})C_{j_1,j_2}(\mathbf{q}^{(j_2)}) \\
- \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}S_{j_1,j_2}(\mathbf{q}^{(j_2)})\big) - q_1^{(j_2)}\frac{\mathbf{u}_0\cdot\mathbf{q}^{(j_2)}}{B_{j_2}(\mathbf{q}^{(j_1)})}\big(E_{j_2}(\mathbf{q}^{(j_1)})S_{j_1,j_2}(\mathbf{q}^{(j_2)}) \\
+ \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}C_{j_1,j_2}(\mathbf{q}^{(j_2)})\big)\Bigg)\Bigg] + 2D_0 t q_1^{(j_2)}[\mathbf{q}^{(j_1)}\cdot\mathbf{p}(\mathbf{q}^{(j_2)})]\,\mathrm{e}^{-E_{j_2}(\mathbf{q}^{(j_1)})t} \\
\times\big(E_{j_2}(\mathbf{q}^{(j_1)})C_{j_1,j_2}(\mathbf{q}^{(j_2)}) - \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}S_{j_1,j_2}(\mathbf{q}^{(j_2)})\big)\Bigg\} \\
- \frac{u_0^2 q_0}{2N}\sum_{j_1}{}'\sum_{j_2}{}'' p_1(\mathbf{q}^{(j_1)})p_1(\mathbf{q}^{(j_2)})\frac{\mathrm{e}^{-t/\tau_D^{(j_1)}}}{B_{j_2}(0)}\big[E_{j_2}(0)C_{j_1,j_2}(0) \\
- \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}S_{j_1,j_2}(0) - \mathrm{e}^{-t/\tau_D^{(j_2)}}\big(E_{j_2}(0)C_{j_1,j_2}(\mathbf{q}^{(j_2)}) - \mathbf{u}_0\cdot\mathbf{q}^{(j_2)}S_{j_1,j_2}(\mathbf{q}^{(j_2)})\big)\big].
\end{aligned}
$$

The following functions are used in this expression: $S_{j_1,j_2}(\mathbf{k}) = \sin\left(\mathbf{u}_0\cdot\mathbf{k}t + t\big/\tau_u^{(j_1)} + \alpha^{(j_1)} + \alpha^{(j_2)}\right)$, $C_{j_1,j_2}(\mathbf{k}) = \cos\left(\mathbf{u}_0\cdot\mathbf{k}t + t\big/\tau_u^{(j_1)} + \alpha^{(j_1)} + \alpha^{(j_2)}\right)$, $B_j(\mathbf{k}) = \left(1\big/\tau_D^{(j)} + 2D_0\mathbf{q}^{(j)}\cdot\mathbf{k}\right)^2 + \left(1\big/\tau_u^{(j)}\right)^2$, and $E_j(\mathbf{k}) = 1\big/\tau_D^{(j)} + 2D_0\mathbf{q}^{(j)}\cdot\mathbf{k}$.

Appendix A.2. Ensemble average of $D_{11}(t)$

From equation (13) we can derive for the ensemble mean of $D_{11}(t)$:

$$
\begin{aligned}
\overline{D_{11}(t)} = D_0 - \frac{1}{2}\partial_{k_1}^2\,\overline{F(\mathbf{k},t)}\Big|_{\mathbf{k}=\mathbf{0}} = D_0 - \frac{1}{2}\partial_{k_1}^2\,\frac{1}{(2\pi)^N}\int_0^{2\pi}\mathrm{d}\alpha^{(1)}\cdots\int_0^{2\pi}\mathrm{d}\alpha^{(N)} \\
\times\frac{l_0^{3N}}{\sqrt{2\pi}^{3N}}\int\mathrm{d}^3q^{(1)}\cdots\int\mathrm{d}^3q^{(N)}\,\mathrm{e}^{-\mathbf{q}^{(1)2}\frac{l_0^2}{2}-\cdots-\mathbf{q}^{(N)2}\frac{l_0^2}{2}}F(\mathbf{k},t)\Big|_{\mathbf{k}=\mathbf{0}}.
\end{aligned}
$$

Substituting $F(\mathbf{k},t)$ and applying $\mathbf{q}\cdot\mathbf{p}(\mathbf{q})=0$, one easily finds

$$\begin{aligned}\overline{D_{11}(t)} &= D_0 - \frac{1}{2}\frac{l_0^{3N}}{\sqrt{2\pi}^{3N}}\int \mathrm{d}^3q^{(1)}\cdots\int \mathrm{d}^3q^{(N)}\,\mathrm{e}^{-\frac{l_0^2}{2}(\mathbf{q}^{(1)^2}+\cdots+\mathbf{q}^{(N)^2})}2\int_0^t \mathrm{d}\tau\sum_j{}'\frac{q_0u_0^2}{2N}p_1^2(\mathbf{q}^{(j)})\\ &\quad\times\left[\mathrm{e}^{-2D_0\mathbf{q}^{(j)^2}t}\,\mathrm{e}^{D_0\mathbf{q}^{(j)^2}\tau+\mathrm{i}\mathbf{u}_0\cdot\mathbf{q}^{(j)}\tau}-\mathrm{e}^{-D_0\mathbf{q}^{(j)^2}\tau+\mathrm{i}\mathbf{u}_0\cdot\mathbf{q}^{(j)}\tau}\right]\\ &= D_0 - q_0u_0^2\frac{l_0^3}{\sqrt{2\pi}^3}\int_0^t \mathrm{d}\tau\int \mathrm{d}^3q\,\mathrm{e}^{-\frac{l_0^2}{2}\mathbf{q}^2}p_1^2(\mathbf{q})\left[\mathrm{e}^{-2D_0\mathbf{q}^2t}\,\mathrm{e}^{D_0\mathbf{q}^2\tau-\mathrm{i}\mathbf{u}_0\cdot\mathbf{q}\tau}-\mathrm{e}^{-D_0\mathbf{q}^2\tau-\mathrm{i}\mathbf{u}_0\cdot\mathbf{q}\tau}\right]\\ &= D_0 + M^-(t/\tau_u,0) - M^+(t/\tau_u,2t/\tau_D)\end{aligned}$$

with

$$M^\pm(T,a) = q_0u_0l_0(2\pi)^{d/2}\int_{\mathbf{k}}\int_0^T \mathrm{d}\tau\,\exp(-(1+2a)\mathbf{k}^2/2)\exp(\pm\epsilon\mathbf{k}^2\tau-\mathrm{i}k_1\tau)\,p_1^2(\mathbf{k}/l_0)$$

for $d=3$ and $\tau_u=l_0/u_0$, $\tau_D=l_0^2/D_0$, $\epsilon=\tau_u/\tau_D$. The function $M^\pm(T,a)$ is found to be (see for $M_1^\pm(T;a,a,a)$ in [8])

$$\begin{aligned}M^\pm(T,a) = \mp q_0u_0l_0\sqrt{\frac{\pi}{2}}\frac{1}{(1+2a)^{(d-1)/2}}\Bigg\{&\operatorname{erf}(g(\mp T))\\ &+\frac{1}{\sqrt{\pi}}\exp(-g^2(\mp T))\left(\frac{1}{g(\mp T)}+4\lambda^2\frac{f(\mp T)}{g^2(\mp T)}-\frac{3}{2g^3(\mp T)}\right)+\operatorname{erf}(g(\mp T))\\ &\times\left(4\lambda^2\frac{f(\mp T)}{g(\mp T)}-\frac{1}{g^2(\mp T)}+\frac{3}{4g^4(\mp T)}-2\lambda^2\frac{f(\mp T)}{g^3(\mp T)}+8\lambda^4\frac{f(\mp T)}{g(\mp T)}\right)\\ &-8\lambda^4\exp\left(\frac{1}{2\lambda^2}\right)\left(\operatorname{erfc}\left(\frac{1}{\sqrt{2}\lambda}\right)-\operatorname{erfc}(f(\mp T))\right)-\frac{4\sqrt{8}}{3\sqrt{\pi}}\lambda-\frac{4\sqrt{8}}{\sqrt{\pi}}\lambda^3\Bigg\}\end{aligned}$$

with $\lambda=\epsilon(1+2a)^{-1/2}$, $f(t)=\frac{(1+2a)\tau_D/\tau_u+t/\tau_u}{\sqrt{2(1+2a)+4t/\tau_D}}$ and $g(t)=\frac{t/\tau_u}{\sqrt{2(1+2a)+4t/\tau_D}}$.

Appendix B. Transport parameters from the iterative solution of the Langevin equation

Appendix B.1. Dispersion coefficient

The explicit expression for $D_{11}(t)$ given by the iteration of the Langevin equation in section 4 reads

$$\begin{aligned}D_{11}(t) = D_0 &+ 2D_0\sqrt{\frac{2q_0}{N}}\sum_j{}^*\,p_1(\mathbf{q}^{(j)})\frac{t}{\tau_u^{(j)}}\,\mathrm{e}^{-t/\tau_D^{(j)}}\sin\left(t/\tau_u^{(j)}+\alpha^{(j)}\right)\\ &-2u_0^2\frac{q_0}{N}\sum_{j_1}{}^*\sum_{j_2}{}^*\,p_1(\mathbf{q}^{(j_1)})\,p_1(\mathbf{q}^{(j_2)})\cos\left(t/\tau_u^{(j_1)}+\alpha^{(j_1)}\right)\frac{\mathrm{e}^{-t/\tau_D^{(j_1)}}}{\left(\tau_D^{(j_2)}\right)^{-2}+\left(\tau_u^{(j_2)}\right)^{-2}}\\ &\times\left[\frac{1}{\tau_D^{(j_2)}}\cos\alpha^{(j_2)}-\frac{1}{\tau_u^{(j_2)}}\sin\alpha^{(j_2)}-\mathrm{e}^{-t/\tau_D^{(j_2)}}\left(\frac{1}{\tau_D^{(j_2)}}\cos\left(t/\tau_u^{(j_2)}+\alpha^{(j_2)}\right)\right.\right.\\ &\left.\left.-\frac{1}{\tau_u^{(j_2)}}\sin\left(t/\tau_u^{(j_2)}+\alpha^{(j_2)}\right)\right)\right]+u_0^2\frac{q_0}{N}\sum_{j_1}{}^*\sum_{j_2}{}^*\,p_1(\mathbf{q}^{(j_1)})\,p_1(\mathbf{q}^{(j_2)})\,\mathrm{e}^{-t/\tau_D^{(j_2)}}\\ &\times\left\{\frac{1}{A^+_{j_1,j_2}}\left[\left(\frac{1}{\tau_D^{(j_1)}}+\frac{2}{\tau_D^{(j_1,j_2)}}\right)\cos\beta^+_{j_1,j_2}(t)-\frac{1}{\tau_u^{(j_1)}}\sin\beta^+_{j_1,j_2}(t)\right.\right.\end{aligned}$$

$$- \mathrm{e}^{-t/\tau_D^{(j_2)} - 2t/\tau_D^{(j_1,j_2)}} \left(\left(\frac{1}{\tau_D^{(j_1)}} + \frac{2}{\tau_D^{(j_1,j_2)}} \right) \cos \gamma^+_{j_1,j_2}(t) - \frac{1}{\tau_u^{(j_1)}} \sin \gamma^+_{j_1,j_2}(t) \right) \Bigg]$$
$$+ \frac{1}{A^-_{j_1,j_2}} \left[\left(\frac{1}{\tau_D^{(j_1)}} - \frac{2}{\tau_D^{(j_1,j_2)}} \right) \cos \beta^-_{j_1,j_2}(t) + \frac{1}{\tau_u^{(j_1)}} \sin \beta^-_{j_1,j_2}(t) - \mathrm{e}^{-t/\tau_D^{(j_2)} + 2t/\tau_D^{(j_1,j_2)}} \right.$$
$$\left. \times \left(\left(\frac{1}{\tau_D^{(j_1)}} - \frac{2}{\tau_D^{(j_1,j_2)}} \right) \cos \gamma^-_{j_1,j_2}(t) + \frac{1}{\tau_u^{(j_1)}} \sin \gamma^-_{j_1,j_2}(t) \right) \right] \Bigg\} \tag{B1}$$

where we used the following definitions and functions: $\sum_j^* := \sum_{j=1}^N$ for $\mathbf{q}^{(j)} \neq \mathbf{0}$, $\tau_D^{(j_1,j_2)} = (D_0 \mathbf{q}^{(j_1)} \mathbf{q}^{(j_2)})^{-1}$, $A^{\pm}_{j_1,j_2} = \left(1/\tau_D^{(j_1)} \pm 2/\tau_D^{(j_1,j_2)}\right)^2 + \left(1/\tau_u^{(j_1)}\right)^2$, $\beta^{\pm}_{j_1,j_2}(t) = t/\tau_u^{(j_2)} + \alpha^{(j_2)} \pm \alpha^{(j_1)}$ and $\gamma^{\pm}_{j_1,j_2}(t) = t/\tau_u^{(j_2)} + \alpha^{(j_2)} \pm \left(t/\tau_u^{(j_1)} + \alpha^{(j_1)}\right)$.

Appendix B.2. Ensemble average of $D_{11}(t)$

The ensemble mean $\overline{D_{11}(t)}$ of $D_{11}(t)$, equation (22), is given by

$$\overline{D_{11}(t)} = D_0 + \int_0^t \mathrm{d}t' \int_{\mathbf{k}} \int_{\mathbf{k}'} \overline{\tilde{w}_1(\mathbf{k}) \tilde{w}_1(\mathbf{k}')}\, \mathrm{e}^{-D_0(\mathbf{k}^2 t + \mathbf{k}'^2 t' + 2\mathbf{k}\cdot\mathbf{k}' t')}\, \mathrm{e}^{-i\mathbf{u}_0\cdot(\mathbf{k}t + \mathbf{k}'t')}$$
$$- \int_0^t \mathrm{d}t' \int_{\mathbf{k}} \int_{\mathbf{k}'} \overline{\tilde{w}_1(\mathbf{k}) \tilde{w}_1(\mathbf{k}')}\, \mathrm{e}^{-D_0(\mathbf{k}^2 t + \mathbf{k}'^2 t')}\, \mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t + \mathbf{k}'t')}.$$

Using $\overline{\tilde{w}_1(\mathbf{k}) \tilde{w}_1(\mathbf{k}')}$ from equation (4), we obtain

$$\begin{aligned}
\overline{D_{11}(t)} &= D_0 + (2\pi)^{d/2} q_0 u_0^2 l_0^d \int_0^t \mathrm{d}t' \int_{\mathbf{k}} p_1^2(\mathbf{k})\, \mathrm{e}^{-\frac{1}{2} l_0^2 \mathbf{k}^2}\, \mathrm{e}^{-D_0(\mathbf{k}^2 t - \mathbf{k}^2 t')}\, \mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t - \mathbf{k}t')} \\
&\quad - (2\pi)^{d/2} q_0 u_0^2 l_0^d \int_0^t \mathrm{d}t' \int_{\mathbf{k}} p_1^2(\mathbf{k})\, \mathrm{e}^{-\frac{1}{2} l_0^2 \mathbf{k}^2}\, \mathrm{e}^{-D_0(\mathbf{k}^2 t + \mathbf{k}^2 t')}\, \mathrm{e}^{-\mathrm{i}\mathbf{u}_0\cdot(\mathbf{k}t - \mathbf{k}t')} \\
&= D_0 + (2\pi)^{d/2} q_0 u_0 l_0 \int_0^{t/\tau_u} \mathrm{d}\tau \int_{\mathbf{k}} p_1^2(\mathbf{k}/l_0)\, \mathrm{e}^{-\frac{1}{2}\mathbf{k}^2}\, \mathrm{e}^{-\epsilon\mathbf{k}^2\tau - \mathrm{i}k_1\tau} \\
&\quad - (2\pi)^{d/2} q_0 u_0 l_0 \int_0^{t/\tau_u} \mathrm{d}\tau \int_{\mathbf{k}} p_1^2(\mathbf{k}/l_0)\, \mathrm{e}^{-\frac{1}{2}\mathbf{k}^2}\, \mathrm{e}^{+\epsilon\mathbf{k}^2\tau - 2\mathbf{k}^2 t/\tau_D - \mathrm{i}k_1\tau} \\
&= D_0 + M^-(t/\tau_u, 0) - M^+(t/\tau_u, 2t/\tau_D)
\end{aligned}$$

where we apply $M^{\pm}(T, a)$ from appendix A.2, and the substitutions $\tau = t'/\tau_u$, $k_i \to k_i/l_0$.

Appendix B.3. Mean square sample-to-sample fluctuations of D_{11}^{∞}

Taking the expression for D_{11}^{∞} of equation (16), we find for $(\delta D_{11}^{\infty})^2$ using the modified distribution function $\tilde{P}_{\mathbf{q}}(\mathbf{q})$:

$$(\delta D_{11}^{\infty})^2 = \overline{(D_{11}^{\infty})^2} - \overline{D_{11}^{\infty}}^2 = \frac{q_0^2 u_0^4}{N^2} \overline{\sum_j^* p_1^4(\mathbf{q}^{(j)}) \frac{D_0^2 \mathbf{q}^{(j)4}}{\left(\left(D_0 \mathbf{q}^{(j)2}\right)^2 + (\mathbf{u}_0 \cdot \mathbf{q}^{(j)})^2\right)^2}} - \frac{1}{N} (\overline{D_{11}^{\infty}} - D_0)^2$$
$$= \frac{q_0^2 u_0^4 D_0^2}{N} \int \mathrm{d}^3 q\, \tilde{P}_{\mathbf{q}}(\mathbf{q}) p_1^4(\mathbf{q}) \frac{\mathbf{q}^4}{((D_0 \mathbf{q}^2)^2 + (\mathbf{u}_0 \cdot \mathbf{q})^2)^2} - \frac{1}{N} (\overline{D_{11}^{\infty}} - D_0)^2$$
$$= 4\pi \frac{q_0^2 u_0^4}{N D_0^2} \int_0^1 \mathrm{d}z (1 - z^2)^4 \int_0^{\infty} \mathrm{d}q\, \tilde{P}_{\mathbf{q}}(\mathbf{q}) \frac{q^2}{(q^2 + (u_0 z/D_0)^2)^2} - \frac{1}{N} (\overline{D_{11}^{\infty}} - D_0)^2.$$

The outer integrations can be performed using Gradshteyn's tables [15]:

$$
\begin{aligned}
(\delta D_{11}^{\infty})^2 &= \frac{q_0^2 u_0^2 l_0^2}{4\epsilon N}\sqrt{\frac{\pi}{2}}\left(\int_{\frac{l_0}{L}}^{1} \mathrm{d}x \frac{1}{x}\exp\left(\frac{x}{2\epsilon^2}\right) - \int_0^1 \mathrm{d}x \frac{1}{x}\exp\left(\frac{x}{2\epsilon^2}\right)\operatorname{erf}\left(\sqrt{\frac{x}{2\epsilon^2}}\right)\right) \\
&\quad + \frac{u_0^2 q_0^2 l_0^2}{N}\left[\sqrt{\frac{\pi}{2}}\,\mathrm{e}^{\frac{1}{2\epsilon^2}}\left(-\frac{\epsilon}{2} - \epsilon^3 + 168\epsilon^7 - 4\epsilon^5\right)\operatorname{erfc}\left(\sqrt{\frac{1}{2\epsilon^2}}\right) + \frac{133}{15}\epsilon^2 + \frac{1}{2}\right. \\
&\quad \left. + 52\epsilon^4 + 168\epsilon^6 - \sqrt{\frac{\pi}{2}}\left(\frac{1}{2\epsilon} + 18\epsilon^3 + 2\epsilon + 80\epsilon^5 + 168\epsilon^7\right)\right] - \frac{1}{N}(\overline{D_{11}^{\infty}} - D_0)^2 \\
&= \frac{q_0^2 u_0^2 l_0^2}{4\epsilon N}\sqrt{\frac{\pi}{2}}\ln(L/l_0) + \frac{G(\epsilon)}{N}
\end{aligned}
$$

where we defined the function $G(\epsilon)$ by

$$
\begin{aligned}
G(\epsilon) &= \frac{u_0^2 q_0^2 l_0^2}{4\epsilon}\sqrt{\frac{\pi}{2}}\left(\sum_{n=1}^{\infty}\frac{\left(\frac{1}{2\epsilon^2}\right)^n}{n!n} - \frac{1}{\sqrt{\pi}}\sum_{n=0}^{\infty}\frac{\sqrt{\frac{1}{2\epsilon^2}}^{2n+1}}{1\times 9\times\cdots\times(2n+1)^2}\right) \\
&\quad + u_0^2 q_0^2 l_0^2\left[\sqrt{\frac{\pi}{2}}\,\mathrm{e}^{\frac{1}{2\epsilon^2}}\left(168\epsilon^7 - 4\epsilon^5 - \epsilon^3 - \frac{\epsilon}{2}\right)\operatorname{erfc}\left(\sqrt{\frac{1}{2\epsilon^2}}\right) + \frac{133}{15}\epsilon^2 + \frac{1}{2}\right. \\
&\quad \left. + 52\epsilon^4 + 168\epsilon^6 - \sqrt{\frac{\pi}{2}}\left(\frac{1}{2\epsilon} + 18\epsilon^3 + 2\epsilon + 80\epsilon^5 + 168\epsilon^7\right)\right] - (\overline{D_{11}^{\infty}} - D_0)^2.
\end{aligned}
$$

The ensemble mean of D_{11}^{∞} is given by

$$
\overline{D_{11}^{\infty}} = D_0 + q_0 u_0 l_0\sqrt{\frac{\pi}{2}}\left(1 - 8\epsilon^4\exp\left(\frac{1}{2\epsilon^2}\right)\operatorname{erfc}\left(\frac{1}{\sqrt{2}\epsilon}\right) + 8\epsilon^4 - \frac{16\epsilon^3}{\sqrt{2\pi}} + 4\epsilon^2 - \frac{16\epsilon}{3\sqrt{2\pi}}\right).
$$

Appendix C. Ensemble averages for the Gaussian white noise

The expressions in section 4 which are averaged over the white noise consist of functional integrals. They arise from the derivation of $D_{11}(t)$, equation (22). Using the definitions of section 2, and $D_{0,ij} = D_0\delta_{ij}$, we obtain from equation (9):

$$
\begin{aligned}
\left\langle \mathrm{e}^{-\mathrm{i}\int_0^t \mathrm{d}s L(s)\xi_1(s)}\right\rangle &= \int \mathcal{D}[\xi_1]\mathcal{P}[\xi_1(s)]\,\mathrm{e}^{-\mathrm{i}\int_0^t \mathrm{d}s L(s)\xi_1(s)} \\
&= \lim_{\Delta\to 0}\int\prod_{i=0}^{N-1}\left(\mathrm{d}\xi_1(s_i)\sqrt{\frac{\Delta}{4\pi D_0}}\,\mathrm{e}^{-\frac{\Delta}{4D_0}(\xi_1(s_i)+2\mathrm{i}D_0 L(s_i))^2}\,\mathrm{e}^{-D_0\Delta L^2(s_i)}\right) \\
&= \lim_{\Delta\to 0}\prod_{i=0}^{N-1}\mathrm{e}^{-D_0\Delta L^2(s_i)} = \mathrm{e}^{-D_0\int_0^t \mathrm{d}s L^2(s)}
\end{aligned}
$$

where $L(s)$ is an arbitrary function. This result can be extended for a vector function $\mathbf{L}(s)$, i.e. $\left\langle \mathrm{e}^{-\mathrm{i}\int_0^t \mathrm{d}s\mathbf{L}(s)\cdot\boldsymbol{\xi}(s)}\right\rangle = \mathrm{e}^{-D_0\int_0^t \mathrm{d}s\mathbf{L}^2(s)}$. For $\mathbf{L}(s) = \mathbf{k}$ we then easily find:

$$
\left\langle \exp\left(-\mathrm{i}\int_0^t \mathrm{d}t'\mathbf{k}\cdot\boldsymbol{\xi}(t')\right)\right\rangle = \exp(-D_0\mathbf{k}^2 t).
$$

For $\mathbf{L}(s) = \mathbf{k}\Theta(t-s) + \mathbf{k}'\Theta(t'-s)$, we obtain

$$\left\langle \exp\left(-\mathrm{i}\mathbf{k}\cdot\int_0^t \mathrm{d}t''\boldsymbol{\xi}(t'') - \mathrm{i}\mathbf{k}'\cdot\int_0^{t'}\mathrm{d}t''\boldsymbol{\xi}(t'')\right)\right\rangle = \left\langle \exp\left(-\mathrm{i}\int_0^t \mathrm{d}s\mathbf{L}(s)\cdot\boldsymbol{\xi}(s)\right)\right\rangle$$
$$= \exp\left(-D_0\int_0^t \mathrm{d}s(\mathbf{L}(s))^2\right) = \exp(-D_0\mathbf{k}^2t - D_0\mathbf{k}'^2t' - 2D_0\mathbf{k}\cdot\mathbf{k}'\min(t,t'))$$

and for $\mathbf{L}(s) = \mathbf{k}\Theta(t'-s)$, we find

$$\left\langle \xi_j(t'')\exp\left(-\mathrm{i}\mathbf{k}\cdot\int_0^{t'}\mathrm{d}t'''\boldsymbol{\xi}(t''')\right)\right\rangle = \mathrm{i}\frac{\delta}{\delta L_j(t'')}\left\langle \exp\left(-\mathrm{i}\int_0^t \mathrm{d}s\mathbf{L}(s)\cdot\boldsymbol{\xi}(s)\right)\right\rangle$$
$$= \mathrm{i}\frac{\delta}{\delta L_j(t'')}\exp\left(-D_0\int_0^t \mathrm{d}s(\mathbf{L}(s))^2\right) = -\mathrm{i}2D_0k_j\Theta(t'-t'')\exp(-D_0\mathbf{k}^2t').$$

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