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S. Attinger · M. Dentz · W. Kinzelbach

Exact transverse macro dispersion coefficients for transport in heterogeneous porous media

Abstract We study transport through heterogeneous media. We derive the exact large scale transport equation. The macro dispersion coefficients are determined by additional partial differential equations. In the case of infinite Peclet numbers, we present explicit results for the transverse macro dispersion coefficients. In two spatial dimensions, we demonstrate that the transverse macro dispersion coefficient is zero. The result is not limited on lowest order perturbation theory approximations but is an exact result. However, the situation in three spatial dimensions is very different: The transverse macro dispersion coefficients are finite – a result which is confirmed by numerical simulations we performed.

Keywords Stochastic modelling · Coarse graining · Dispersion · Transport · Heterogeneous porous media

1 Introduction

Asymptotically, plumes moving in single-scale heterogeneous porous media are efficiently modelled by transport models with equivalent large scale or macro transport parameters. The large scale dispersion tensor is anisotropic even if the conductivity tensor of the medium is isotropic. The anisotropy of the dispersion tensor stems from the fact that the spreading of the plume is larger in the direction of the flow field than in transverse directions.

In general, the perturbation theory approach has been proved to be a valuable method for the quantitative prediction of transport properties in heterogeneous porous media. For a comprehensive review the reader is referred to the textbooks of (Dagan, 1989) and (Gelhar, 1993). In lowest order perturbation theory, the longitudinal macro dispersion coefficient increases by several orders of magnitude, whereas the transverse only approaches an asymptotic value in the order of magnitude of the pore-scale coefficient. In media of moderate heterogeneity, therefore, the transverse mixing of compounds is hardly enhanced. The longitudinal dispersion coefficient derived in the lowest order approach fits very well with field findings (Freyberg, 1986), whereas the transverse macro dispersion coefficient underestimates the experimental data by at least one order of magnitude (Gelhar and Axness, 1983).

However, transverse mixing is one of the critical factors in natural attenuation processes. A remedial action involves the injection of the adsorbing organic pollutant (e.g. chlorinated sorbents, hydrocarbons), an electron acceptor (e.g. oxygen) and the degrading microbial population. All chemical compounds have to be available at the same time and at the same location. One of the major limitations is the amount of ambient oxygen present in the plume (Alvarez-Cohen, 1993; Thomas and Ward, 1989; Sims et al., 1992). Field results (Freyberg et al., 1986; Moltyaner and Killey, 1988) have shown that transport of oxygen into the plume is dominated by lateral mixing. Therefore, a small lateral or transverse mixing coefficient becomes an important rate-limiting factor in biodegradation (Cirpka and Kitanidis, 2000; Thornton et al., 2001; Thullner et al., 2002).

Motivated by these open questions, Dagan (1994) investigated corrections to the transverse dispersion coefficient in two dimensions due to fourth order perturbation theory contributions in the limit of high Peclet numbers. Asymptotically, these contributions turned out

S. Attinger (✉)
Computational Laboratory (CoLab),
Swiss Federal Institute of Technology (ETH),
8092 Zurich, Switzerland
e-mail: sabine.attinger@env.ethz.ch

M. Dentz
Department of Geotechnical Engineering and
Geosciences Technical University of Catalonia
(UPC) Barcelona, Spain

W. Kinzelbach
Institute of Hydromechanics and Water Resources
Management (IHW)
Swiss Federal Institute of Technology (ETH),
Zurich, Switzerland

to be zero. Hsu et al. (1996) generalized this two dimensional analysis by taking into account also corrections to the linearized Darcy flow field by means of second order terms in the log hydraulic conductivity. The resulting contributions to the dispersion coefficients, however, yielded only minor corrections to the second order results.

The central aim of this paper is to overcome the limitations of perturbation theory approaches in the determination of large scale dispersion coefficients. We make use of a filtering method called Coarse Graining (McComb, 1990) and derive exact expressions for the large scale dispersion coefficients.

2 Heterogeneous transport model

A solute plume released in an aquifer is transported by advection and spreads with time due to dispersion. Its movement is modelled by

$$n\partial_t c(\mathbf{x},t) + \nabla\cdot(\mathbf{u}(\mathbf{x}) - \mathbf{D}_0\nabla)c(\mathbf{x},t) = \rho_c(\mathbf{x})\delta(t) \qquad (1)$$

where $c(\mathbf{x},t)$ is the spatial concentration of the solute. Here and in the following, we denote vector and tensor quantities by boldface characters. The flow field $\mathbf{u}(\mathbf{x})$ follows from Darcy's Law

$$\mathbf{u}(\mathbf{x}) = -K_f(\mathbf{x})\nabla\phi(\mathbf{x}) \qquad (2)$$

where $\phi(\mathbf{x})$ is the piezometric head and $K_f(\mathbf{x}) = K_g\exp(\tilde{f})$ a local conductivity field. K_g is the geometric mean and $\tilde{f}$ is a normal distributed random field with vanishing mean. The mean flow field is uniform and aligned in $\mathbf{e}_1$-direction, $\bar{u}\mathbf{e}_1$. The tensor $\mathbf{D}_0$ is the local dispersion tensor. n is the porosity. As boundary condition we assume vanishing concentrations at infinity. The initial concentration distribution is given by $\rho_c(\mathbf{x})$. It is assumed to be spatially extended over many correlations lengths of the log conductivity in transverse directions.

3 Method of Coarse Graining

The aim of applying Coarse Graining is to average local functions over volumes of intermediate size in order to obtain smoothed functions on coarser resolution scales. Originally, this idea was developed in the theory of turbulence (McComb, 1980). In groundwater research, Beckie (2001) and Dykaar and Kitanidis (1992) made use of this approach for studying groundwater flows, Rubin (1999) for investigating solute transport in heterogeneous media. They combined Coarse Graining with a perturbation theory analysis that limited again the results to moderately heterogeneous media.

The coarse scale can be any resolution scale of a measurement or discretization scale of a numerical simulation [Beckie, 2001]. In the following, we assume that the coarse scale characterized by the length λ is much larger than the correlation length of the heterogeneous medium because – in this paper- we focus on the asymptotic behaviour. In other words, we perform the spatial average over a representative volume of the heterogeneous medium. Moreover, the averaging volume must not only be large compared to the velocity correlation scale but also small in comparison to the scale of the plume. Nevertheless, the applicability of Coarse Graining in not limited to the average over representative volumes but applies to the average over smaller volumes as well.

Fluctuations are smoothed out over a typical volume λ^d around the location $\mathbf{x}$ by the following averaging procedure

$$\langle c(\mathbf{x},t)\rangle_\lambda \equiv \frac{1}{\lambda^d}\int_{\mathbf{x}'\in\lambda^d} d^d x c(\mathbf{x}+\mathbf{x}',t) \qquad (3)$$

where $\langle c(\mathbf{x},t)\rangle_\lambda$ is the coarser concentration distribution, d the spatial dimension and $d^d x$ an infinitesimal volume element. Because we focus on the asymptotic limit only, the averaged concentration is called $\langle c(\mathbf{x},t)\rangle_\infty$. The smoothing procedure in (3) can be also viewed as a convolution of the function $c(x,t)$ with a Heavyside function. Transforming the convolution into Fourier space yields a product of the Fourier transform $\hat{c}(k,t)$ with a function that filters out Fourier modes k that are larger than a cut-off value $\propto \lambda^{-1}$ (Rubin, 1999).

We apply Coarse Graining to the heterogeneous transport model (1) and derive the exact large scale transport model in appendix A.

4 Large scale transport model

The exact large scale transport behaviour is described by

$$n\partial_t\langle c(\mathbf{x},t)\rangle + \bar{\mathbf{u}}\cdot\nabla\langle c(\mathbf{x},t)\rangle - \nabla\cdot(\mathbf{D}_0 + \delta\mathbf{D}^{\text{macro}})\nabla\langle c(\mathbf{x},t)\rangle = 0 \qquad (4)$$

with a constant macro dispersion tensor, $D_0 + \delta\mathbf{D}^{\text{macro}}\cdot\langle c\rangle$ denotes the large scale concentration distribution. The unresolved subscale effects are now modelled by the tensor $\delta\mathbf{D}^{\text{macro}}$ with the entries

$$\delta D_{ij}^{\text{macro}} \equiv \langle\tilde{u}_i\chi_j\rangle = \overline{\tilde{u}_i\chi_j} \qquad (5)$$

The auxiliary fields χ_j solve

$$(\mathbf{u}(\mathbf{x})\cdot\nabla - \nabla\mathbf{D}_0\nabla)\chi_j = \tilde{u}_j \qquad (6)$$

$\tilde{u}_j$ denotes the fluctuating part of the heterogeneous velocity component $u_j(\mathbf{x})$. Making use of the ergodicity assumption, we may replace in (5) the spatial average $\langle\cdots\rangle$ by the ensemble average $\overline{(\ldots)}$. Our large scale transport equation equals the transport equation of Neuman and Zhang (1990), Zhang and Neuman (1990) and Guardagnini and Neuman (2001) after localization. The large scale transport equation is well known in context of two-scale homogenisation theory. Lunati et al. (2002) give a detailed derivation for it.

In many realistic field cases, the Peclet number, $\text{Pe} = \bar{u}l/D_0$, is large, $\text{Pe} \gg 1$, with l the correlation length of the log-permeability field. Hence, the set of PDE's (5) can be approximated by

$$\mathbf{u}(\mathbf{x})\cdot\nabla\chi_j(\mathbf{x}) = \tilde{u}_j(\mathbf{x}) \qquad (7)$$

In other words, we focus on purely advective transport phenomena in heterogeneous porous media.

5 Second order results for the transverse macro dispersion coefficients

To obtain lowest or second order results, we split the velocity field in Eq. (7) into the large scale part $\bar{u}$ and its deviation $\tilde{\mathbf{u}}(\mathbf{x})$. We collect the fluctuating parts of the velocity field on the right hand side of (7) and truncate the perturbation series for χ_j by avoiding the second term

$$\bar{u}\partial_{x_1}\chi_j(x) = \tilde{u}_j(x) \tag{8}$$

In second order, the transverse dispersion coefficients then read

$$\begin{aligned} D_{22}^{\text{macro}} &= \overline{\tilde{u}_2\chi_2} = \frac{1}{\bar{u}}\int_{-\infty}^{x_1} \mathrm{d}x_1' \overline{\tilde{u}_2(x_1,x_2,x_3)\tilde{u}_2(x_1',x_2,x_3)} \\ &= \frac{1}{\bar{u}}\int_0^{\infty} \mathrm{d}x_1' \overline{\tilde{u}_2(x_1',0,0)\tilde{u}_2(0,0,0)} \end{aligned} \tag{9}$$

using translation invariance of the velocity correlations in the last manipulation. x_1, x_2 and x_3 denote the components of the spatial vector $\mathbf{x}$. D_{33}^{macro} follows analogously. Moreover, the velocity correlations are even in x_1 and we can write

$$D_{22}^{\text{macro}} = \frac{1}{2\bar{u}}\int_{-\infty}^{\infty} \mathrm{d}x_1' \overline{\tilde{u}_2(x_1',0,0)\tilde{u}_2(0,0,0)} \tag{10}$$

The integral scale of the transverse velocity correlations is zero (Hole-effect) and the transverse dispersion coefficients always vanish in second order approximation. Why do transverse velocity correlations show the Hole-Effect whereas the longitudinal not? An explanation for the transverse integrals scale being equal to zero can be obtained by making use of the potentials generating the flow field.

5.1 Transverse integral scale in two dimensions

In two dimensions, it always exists a potential φ called Lagrange stream function (Bear, 1972) such that for any incompressible flow field with components $(\tilde{u}_1, \tilde{u}_2)$

$$\partial_{x_1}\varphi(x) = \tilde{u}_2(x) \text{ and } \partial_{x_2}\varphi(x) = -\tilde{u}_1(x) \tag{11}$$

holds. Accordingly to the boundary conditions of the flow field, the stream function vanishes at infinity as well. Making use of this two dimensional stream function, it immediately follows that the integral scale of the any transverse velocity correlations in two dimensions-and in turn the transverse macro dispersion coefficient-must be zero

$$\begin{aligned} D_{22}^{\text{macro}} &= \frac{-1}{2\bar{u}}\int_{-\infty}^{+\infty} \mathrm{d}x_1 \partial_{x_1}^2 \overline{\varphi(x_1,0)\varphi(0,0)} \\ &= \frac{-1}{2\bar{u}}\partial_{x_1}\overline{\varphi(x_1,0)\varphi(0,0)}\Big|_{x_1=-\infty}^{x_1=+\infty} = 0 \ . \end{aligned} \tag{12}$$

Evidently, the integral scale of the longitudinal velocity correlations is always finite.

5.2 Transverse integral scales in three dimensions

In three dimensions, the set of equations (8) describes a more complicated relationship. If two potentials φ and φ' exist such that

$$\partial_{x_1}\varphi(x) = \tilde{u}_2(x) \quad \partial_{x_1}\varphi'(x) = \tilde{u}_3(x) \tag{13}$$

holds and both potentials go to zero at infinity, it implies again zero transverse integral scales of the velocity correlations. A very important point is that flow potentials of the form of (13) do not describe the most general three dimensional flow. In fact, the existence of such potentials is equivalent with a vanishing rotation of the flow field in mean flow direction $\mathbf{e}_1$.

Proof. Without any restrictions, any incompressible flow field can be derived from a vector potential $\mathbf{A}(\mathbf{x})$ according to

$$\mathbf{u}(\mathbf{x}) = \nabla \times \mathbf{A}(\mathbf{x}) \tag{14}$$

Moreover, we have the freedom to set $\nabla \cdot \mathbf{A}(\mathbf{x}) = 0$ (Coulomb gauge) in order to guarantee the uniqueness of the vector potential. A representation of the flow field as given in (13) is not the most general one but implies a vector potential of the following form

$$\begin{pmatrix} A_1 \\ A_2 \\ A_3 \end{pmatrix} = \begin{pmatrix} 0 \\ \varphi' \\ -\varphi \end{pmatrix} \tag{15}$$

In other words, the first component of the vector potential A_1 has to be zero. Next, we apply the rotation to (14) that yields together with the Coulomb gauge

$$\nabla \times \mathbf{u}(\mathbf{x}) = \nabla \times \nabla \times \mathbf{A}(\mathbf{x}) = \Delta\mathbf{A}(\mathbf{x}) \tag{16}$$

For vanishing A_1, it directly follows that the rotation of the flow field in mean flow direction $[\nabla \times \mathbf{u}(\mathbf{x})]_1$ vanishes as well. ■

Summarized, in two dimensions the transverse integral scale of the velocity correlations always vanishes whereas in three dimensions the transverse integral scales only vanish for flow fields with vanishing rotation in mean flow direction. In particular, Darcy's flow approximated up to second order in the log conductivity field (linearized version) shows vanishing rotation in mean flow direction. Therefore, the transverse macro dispersion coefficients are zero in second order approximation.

In the next sections, we overcome the limitations of perturbation theory and derive exact results for the transverse dispersion coefficients.

6 Exact transverse macro dispersion coefficient in two dimensions

We look for the exact solution for the field χ_2 (7) to insert it afterwards into expression (6). Our main finding in this section will be that the exact solution is equal to its second order approximation

$$\chi_2(\mathbf{x}) = \frac{1}{\bar{u}}\int^{x_1} \mathrm{d}x_1'\tilde{u}_2(x_1', x_2) \text{ or } \bar{u}\partial_{x_1}\chi_2(\mathbf{x}) = \tilde{u}_2(\mathbf{x}) \quad (17)$$

We will give a strict proof for this statement in the following. As already discussed in Sec. 5.1., (17) implies that the field χ_2 is the stream function of the fluctuating velocity field $\tilde{\mathbf{u}}$. For every stream function, the gradient of the stream function is orthogonal to the velocity field and therefore $\tilde{\mathbf{u}}(\mathbf{x}) \cdot \nabla\chi_2 = 0$ holds. This also follows by simply inserting the solution (17) into (7)

$$\begin{aligned}\tilde{u}_1\partial_{x_1}\chi_2 + \tilde{u}_2\partial_{x_2}\chi_2 &= \tilde{u}_1\frac{\tilde{u}_2}{\bar{u}} + \tilde{u}_2\partial_{x_2}\frac{1}{\bar{u}}\int^{x_1} \mathrm{d}x_1'\tilde{u}_2(x_1', x_2)\\ &= \tilde{u}_1\frac{\tilde{u}_2}{\bar{u}} - \frac{\tilde{u}_2}{\bar{u}}\tilde{u}_1 = 0\end{aligned}$$

The manipulation makes use of the property that the Darcy flow field is divergence free, $\partial_{x_1}\tilde{u}_1 = -\partial_{x_2}\tilde{u}_2$.

Our theoretical result is supported by numerical simulations based on a random walk method. The simulations are performed using 2000 realizations of a Darcy flow field that is approximated by a simplified linearized version (Dentz et al., 2002). The value of the mean flow field is normalized to one and the Peclet number is 1000. The behaviour of the transverse macro dispersion coefficient depending on the variance of the log-conductivity values is plotted in Fig. 1.

The numerical result supports our finding that the exact solution for χ_2 equals its second order approximation (dashed line). All higher order perturbation theory contributions seem to cancel out. Accounting for perturbation theory contributions up to fourth order, this has been also shown by Dagan (1994). Our result is exact in all higher orders perturbation theory.

Moreover, our result demonstrates that using Corrsin's conjecture (dotted line) yields inconsistent results. Corrsin's conjecture (Dagan, 1988) is a self-consistent approximation that replaces the full Greens function in (A7) by the Greens function of the large scale transport equation. It results an implicit set of equations for the macro dispersion coefficients that is consistent with lowest order results. Corrsin's conjecture takes some but not all higher order perturbation theory contributions into account. Unfortunately, the neglected terms are essential to get the correct result.

7 Transverse macro dispersion coefficients in three dimensions

Encouraged from the results for the transverse macro dispersion coefficient found in two dimensions, we now investigate the system of PDE's (7) in three dimensions,

$$\mathbf{u}(\mathbf{x}) \cdot \nabla\chi_2(\mathbf{x}) = \tilde{u}_2(\mathbf{x}) \quad \mathbf{u}(\mathbf{x}) \cdot \nabla\chi_3(\mathbf{x}) = \tilde{u}_3(\mathbf{x}) \quad (18)$$

The simplest idea is to test if – similar to the two dimensional case – the second order solutions

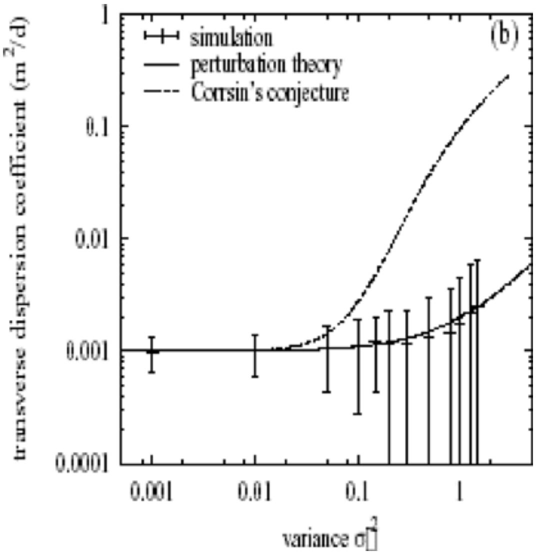

Fig. 1 Transverse Macro Dispersion Coefficient against σ_f^2 in two dimensions (Dentz et al., 2003)

$$\bar{u}\partial_{x_1}\chi_2(\mathbf{x}) = \tilde{u}_2(\mathbf{x}) \quad \bar{u}\partial_{x_1}\chi_3(\mathbf{x}) = \tilde{u}_3(\mathbf{x}) \quad (19)$$

solve the set of Eq. (18). Following this idea, we insert the solutions (19) into the set of Eq. (18) and find that they **only** solve the Eq. (10) **if** additionally $\nabla\chi_2$ and $\nabla\chi_3$ are linearly dependent vectors.

Proof. The set of Eq. (19) is equivalent to

$$\tilde{\mathbf{u}}(\mathbf{x}) \cdot \nabla\chi_2 = 0 \quad \tilde{\mathbf{u}}(\mathbf{x}) \cdot \nabla\chi_3 = 0 \quad (20)$$

Applying representation (19) for the transverse velocity components, we get

$$\begin{aligned}\bar{u}\partial_{x_1}\chi_3\partial_{x_2}\chi_2 = \tilde{u}_3(\mathbf{x})\partial_{x_2}\chi_2 = \tilde{u}_2(\mathbf{x})\partial_{x_2}\chi_3 = \bar{u}\partial_{x_1}\chi_2\partial_{x_2}\chi_3\\ \bar{u}\partial_{x_1}\chi_3\partial_{x_3}\chi_2 = \tilde{u}_3(\mathbf{x})\partial_{x_3}\chi_2 = \tilde{u}_2(\mathbf{x})\partial_{x_3}\chi_3 = \bar{u}\partial_{x_1}\chi_2\partial_{x_3}\chi_3\end{aligned} \quad (21)$$

or in vector form $\nabla\chi_2 \times \nabla\chi_3 = 0$ which implies that $\nabla\chi_2$ and $\nabla\chi_3$ have to be linearly dependent vectors,

$$\nabla\chi_2(\mathbf{x}) = a(\mathbf{x})\nabla\chi_3(\mathbf{x}) \quad \text{with } a(\mathbf{x}) = \frac{\tilde{u}_2(\mathbf{x})}{\tilde{u}_3(\mathbf{x})} \ . \quad (22)$$

$a(\mathbf{x})$ follows by using (19).

By means of equations (20), $\nabla\chi_2$ and $\nabla\chi_3$ are proportional to the gradients of the three dimensional stream functions of the flow field $\tilde{\mathbf{u}}(\mathbf{x})$ (Bear, 1972). In general, the gradients of the three dimensional stream functions together with the flow field $\tilde{\mathbf{u}}(\mathbf{x})$ are assumed to form a three dimensional coordinate system. Hence, the directions of $\nabla\chi_2$ and $\nabla\chi_3$ must not collapse to one direction, the Eq. (18) and (19) can not be valid at the same time and the lowest order solutions (19) do not solve the set of Eq. (18). This heuristic proof is complemented by the correct mathematical proof listed in appendix B.

In fact, transverse macro dispersion coefficients in three dimensions are not only determined by second order terms that give zero contribution. Higher order contributions do not cancel out yielding finite transverse macro dispersion coefficients in three dimensions.

Again, we tested our theoretical result by numerical simulations of particle transport in linearized Darcian flow fields. Linearized Darcian flow fields show a vanishing rotation in $\mathbf{e}_1$-direction. The numerical simulation yields transverse macro dispersion coefficients that increase with increasing variance of the log-permeability field as plotted in Fig. 2. The numerical values clearly differ from the results found in lowest order perturbation theory. Moreover, we realize that using Corrsin's conjecture in three dimensions (dotted line) better reproduces the behaviour of the transverse macro dispersion coefficient than in two dimensions. The reason is that the higher order terms do not cancel exactly. Corrsin's conjecture does not exactly account for all their contributions but approximates them reasonably well. Based on a quasilinear version of Corrsin's conjecture, the first researchers who have demonstrated that transverse dispersivity in 3D goes asymptotically to a nonzero constant were Neuman and Zhang (1990) and Zhang and Neuman (1990).

8 Conclusions

In this article, we studied macro dispersion coefficients for transport in flow fields that are uniform in the mean. We derived the exact large scale transport equation without making use of a perturbation theory approximation. Thus, our results are not limited to moderately heterogeneous media. For transport with infinite Peclet numbers, we gave a strict proof that the transverse macro coefficient in two dimensions is **zero** for transport in steady state divergence free flow fields. The strict proof is a novelty in the literature. In contrast, the transverse macro dispersion coefficients in three dimensions have been proved to be **finite**. The reason may be that in three-dimensional domains flow lines can twiggle and pass each other without intersecting. Therefore, solute particles that are in transversal direction close to each other at some time can be found far apart from each other at later times. For comparison, in two dimensions flow lines can never pass each other and particles cannot move far apart from each other in transverse direction.

Our findings demonstrate a fundamental difference for transport in two and three dimensions. It implies for practical applications that one should be careful modelling three dimensional transport by means of two-dimensional models. A two dimensional situation might underestimate the transverse mixing considerably.

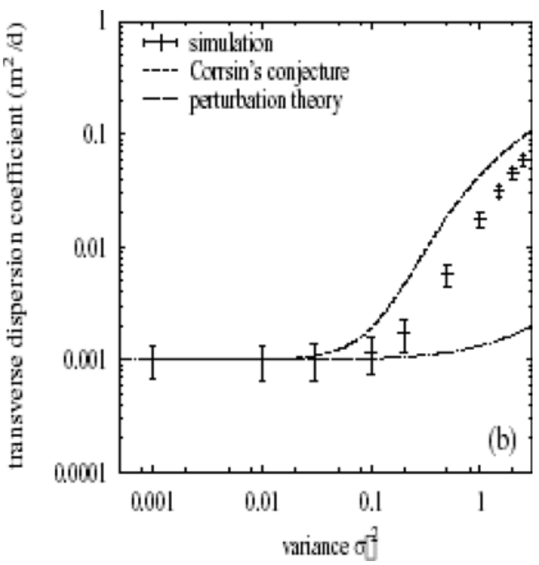

Fig. 2 Transverse Macro Dispersion Coefficient against σ_f^2 in three dimensions (Dentz et al., 2002)

Appendix A

Transforming (3) into the Fourier Space yields a product of the Fourier transform $\hat{c}(k,t)$ with a function $\mathbf{P}^{<}$ that filters out Fourier modes k that are larger than a cut-off value $\propto \lambda^{-1}$. In the following, the filtered concentration in the Fourier Space is called $\hat{c}^{<}$. This equivalence can be used for establishing a systematic Coarse Graining procedure. For a more detailed introduction into the concept of Coarse Graining, we refer to the book of McComb (1990) and the review article of Bouchaud and Georges (1990).

Step 1. In the normal space, we now write the transport equation in operator notation

$$\mathbf{L}c = \rho_c \tag{A1}$$

where $\mathbf{L}$ denotes the transport operator $\partial_t + \mathbf{u}(\mathbf{x})\cdot\nabla - \nabla\mathbf{D}_0\nabla$. We split the operator $\mathbf{L}$ into its mean, $\mathbf{L}^0 = \partial_t + \bar{\mathbf{u}}\cdot\nabla - \nabla\mathbf{D}_0\nabla$, and the deviation from this operator, $\tilde{\mathbf{L}} = \tilde{\mathbf{u}}(\mathbf{x})\cdot\nabla$. Both operators are transformed into Fourier space denoted as $\hat{\mathbf{L}}, \hat{\tilde{\mathbf{L}}}$. The filtered operators are denoted as $\hat{\mathbf{L}}^{<}, \hat{\tilde{\mathbf{L}}}^{<}$.

Step 2. Employing the filter $\mathbf{P}^{<}$ to the Fourier transformed transport equation, an equation for $c^{<}$ can be derived,

$$\hat{\mathbf{L}}^{<}c^{<} = \hat{\rho}_c^{<} - \hat{\tilde{\mathbf{L}}}^{<}c^{>} \tag{A2}$$

The Fourier Back transformation of this equation will yield the desired equation for the smoothed concentration field.

Step 3. By employing also the complementary operator $1 - \mathbf{P}^< \equiv \mathbf{P}^>$, another equation follows for $c^>$,

$$\hat{\mathbf{L}}^> c^> = \hat{\rho}_c^> - \hat{\tilde{\mathbf{L}}}^> c^< \tag{A3}$$

Both equations are coupled.

Step 4. Solving (A3) for $c^>$ and inserting the solution into (A2) we obtain an expression for $c^<$ from which $c^>$ has been eliminated. For spatially extended initial concentrations, $\rho_c^> = 0$ holds and $c^>$ might be written as

$$c^> = (\hat{\mathbf{L}}^>)^{-1} \hat{\tilde{\mathbf{L}}}^> c^< \tag{A4}$$

with the inverse operator $(\hat{\mathbf{L}}^>)^{-1}$ that actually corresponds with the Green's Function of the subscale model. We insert (A4) into (A2) for closure of equation (A2).

Step 5. Finally, the Fourier Back Transformation of (A2) in combination with (A4) gives the desired equation for the smoothed field $\langle c(x,t) \rangle_\infty$. Especially, employing localization of the nonlocal dispersive flux (Neuman, 1993) the smoothed transport model follows as

$$\mathbf{L}^{CG} \langle c \rangle_\infty = \langle \rho_c \rangle_\infty \tag{A5}$$

with a coarse-grained operator $\mathbf{L}^{CG}$ defined by

$$\mathbf{L}^{CG} \equiv \mathbf{L}^0 + \langle \widetilde{\mathbf{L}} (\mathbf{L})^{-1} \widetilde{\mathbf{L}} \rangle_\infty \tag{A6}$$

The first part is the heterogeneity independent part of the transport operator, $\mathbf{L}^0$. The second part of the operator accounts for unresolved subscale effects. Moreover, in the asymptotic limit, the Greens function $\mathbf{L}^{-1}$ reduces to the inverse of the steady state transport operator $\mathbf{L}_\infty = \mathbf{u}(\mathbf{x}) \cdot \nabla - \nabla \mathbf{D}_0 \nabla$. The second part of the smoothed operator is the most interesting part. Thus, we state the entries of this tensor operator in explicit form

$$\partial_{x_i} \delta \mathbf{D}_{ij}^{\text{macro}} \partial_{x_j} \equiv \partial_{x_i} \left\langle \int \mathrm{d}^d x' \tilde{\mathbf{u}}(\mathbf{x})_i \mathbf{L}^{-1}(\mathbf{x}, \mathbf{x}') \tilde{u}(\mathbf{x}')_j \right\rangle_\infty \partial_{x_j} \tag{A7}$$

For shorter notation, we define $\int \mathrm{d}^d x' \mathbf{L}^{-1}(\mathbf{x}, \mathbf{x}') \tilde{u}(\mathbf{x}')_j \equiv \chi_j$.

Appendix B

In the appendix, we present the detailed proof of our statement that there exists no three dimensional flow field that shows a vanishing rotation in $\mathbf{e}_1$-direction and additionally can be derived by a vector potential of the form (13) with the components χ_2 and χ_3 whose gradients are linearly dependent.

In a preliminary step, we will show that

$$\nabla^2 \chi_2(\mathbf{x}) = a(\mathbf{x}) \nabla^2 \chi_3(\mathbf{x}) \tag{B1}$$

holds.

Proof.

$$\bar{u} \nabla^2 \chi_3 = \nabla^2 A_2 = -[\nabla \times \mathbf{u}(\mathbf{x})]_2 = -[\nabla \tilde{f} \times \tilde{\mathbf{u}}(\mathbf{x})]_2 - \bar{u} \partial_{x_3} \tilde{f}$$
$$= \tilde{u}_3 \partial_{x_1} \tilde{f} - \tilde{u}_1 \partial_{x_3} \tilde{f} - \bar{u} \partial_{x_3} \tilde{f}$$

and analogously

$$\bar{u} \nabla^2 \chi_2 = -\nabla^2 A_3 = [\nabla \times \mathbf{u}(\mathbf{x})]_3 = [\nabla \tilde{f} \times \tilde{\mathbf{u}}(\mathbf{x})]_3 - \bar{u} \partial_{x_2} \tilde{f}$$
$$= \tilde{u}_2 \partial_{x_1} \tilde{f} - \tilde{u}_1 \partial_{x_3} \tilde{f} - \bar{u} \partial_{x_2} \tilde{f}$$

Together with $\partial_{x_2} \tilde{f} = a(x) \partial_{x_3} \tilde{f}$ which can be derived directly from $[\nabla \times \mathbf{u}(\mathbf{x})]_1 = 0$ (B1) follows. ■

Next, we write $\nabla^2 \chi_2 = \nabla(a(x) \nabla \chi_3) = a(x) \nabla^2 \chi_3 + \nabla a(x) \cdot \nabla \chi_3$. Therefore, the last term has to be zero. In other words, $\nabla a(x)$ has to be orthogonal to $\nabla \chi_3$. However, this contradicts with

$$\nabla \times \nabla \chi_2 = \nabla \times (a(x) \nabla \chi_3) = \nabla a(x) \times \nabla \chi_3 = 0$$

which implies that $\nabla a(x)$ is parallel to $\nabla \chi_3$. ■

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