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ASYMPTOTICS OF SOLUTE DISPERSION IN PERIODIC POROUS MEDIA*

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Abstract. The concentration $C(\mathbf{x}, t)$ of a solute in a saturated porous medium is governed by a second-order parabolic equation $\partial C/\partial t = -U_0\mathbf{b}\cdot\nabla C + \frac{1}{2}\sum D_{ij}\partial^2 C/\partial x_i\,\partial x_j$. In the case that $\mathbf{b}$ is periodic and divergence free, and D_{ij} are constants and $((D_{ij}))$ positive definite, the concentration is asymptotically Gaussian for large times. This article analyzes the dependence of the dispersion matrix $\mathbf{K}$ of the limiting Gaussian distribution on the velocity parameter U_0 and the period "a." It is shown that each coefficient K_{ii} is asymptotically quadratic in aU_0 if $b_i - \bar{b}_i$ has a nonzero component in the null space of $\mathbf{b}\cdot\nabla$, and asymptotically constant in aU_0 if $b_i - \bar{b}_i$ belongs to the range of $\mathbf{b}\cdot\nabla$. It is shown in a more general context that $\mathbf{K}$ depends only on aU_0. An asymptotic expansion of the Cramer–Edgeworth type is derived for concentration refining the Gaussian approximation.

Key words. Markov process, large scale dispersion, eigenfunction expansion, singular perturbation, range, null space

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1. Introduction. Consider a nonreactive dilute solute injected into a porous medium saturated with a liquid under nonturbulent flow. Suppose the following parabolic equation governing solute concentration $C(\mathbf{x}, t)$ at position $\mathbf{x}$ at time t holds at a certain space-time scale:

$$\frac{\partial C}{\partial t} = \frac{1}{2}\sum_{i,j=1}^{n} \frac{\partial^2}{\partial x_i\,\partial x_j}\left(D_{ij}\left(\frac{\mathbf{x}}{a}\right)C\right) - \sum_{i=1}^{n}\frac{\partial}{\partial x_i}\left(U_0 b_i\left(\frac{\mathbf{x}}{a}\right)C\right),\qquad \mathbf{x} = (x_1, x_2, \cdots, x_n) \in \Re^n, \quad t > 0. \tag{1.1}$$

In (1.1), $U_0\mathbf{b}(\mathbf{x}/a) = U_0(b_1(\mathbf{x}/a), b_2(\mathbf{x}/a), \cdots, b_n(\mathbf{x}/a))$ denotes the solute drift velocity vector, $D(\mathbf{x}/a) = ((D_{ij}(\mathbf{x}/a)))$ is a positive-definite symmetric matrix, and U_0, a are positive scalars. The parameters U_0 and a scale liquid velocity and spatial length, respectively. Although in the physical context $n = 3$, for mathematical purposes we let n be arbitrary.

The solution $C(\mathbf{x}, t)$ of (1.1) is given by (Friedman (1975, pp. 139–144)),

$$C(\mathbf{x}, t) = \int_{\Re^n} h(\mathbf{z})p(t; \mathbf{z}, \mathbf{x})\,d\mathbf{z}, \tag{1.2}$$

where h is the continuous, bounded, initial concentration, and $p(t; \mathbf{z}, \mathbf{x})$ is the fundamental solution of (1.1). Conditions on the coefficients $b_i(\mathbf{x})$, $D_{ij}(\mathbf{x})$ that guarantee the uniqueness and necessary smoothness of the fundamental solution are assumed throughout. Now $p(t; \mathbf{z}, \mathbf{x})$ is also the transition probability density function of the Markov process $\mathbf{X}(t)$ defined by Itô's stochastic differential equation

$$d\mathbf{X}(t) = U_0\mathbf{b}(\mathbf{X}(t)/a)\,dt + \boldsymbol{\sigma}(\mathbf{X}(t)/a)\,d\mathbf{B}(t),\qquad \mathbf{X}(0) = \mathbf{z}, \tag{1.3}$$

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where $\sigma(\mathbf{x})$ is the positive-definite matrix the square of which is $\mathbf{D}(\mathbf{x})$ and $\mathbf{B}(t) = (B_1(t), B_2(t), \cdots, B_n(t))$ is an n-dimensional standard Brownian motion process.

Analyzing the asymptotic behavior of $C(\mathbf{x}, t)$ for large t is equivalent to analyzing the asymptotic behavior of $\mathbf{X}(t)$ for large t. To be more specific, suppose that the stochastic process

$$\mathbf{Z}_\varepsilon(t) \equiv \varepsilon[\mathbf{X}(t/\varepsilon^2) - \varepsilon^{-2} U_0 \bar{\mathbf{b}} t] \tag{1.4}$$

converges in distribution, as $\varepsilon \downarrow 0$, to a Brownian motion with zero mean and a dispersion matrix $\mathbf{K} = ((K_{ij}))$. Here $\bar{\mathbf{b}} = (\bar{b}_1, \cdots, \bar{b}_n)$ is a suitable constant vector interpreted as the *large scale average* of $\mathbf{b}(\mathbf{x})$. In other words, suppose that a *central limit theorem* (CLT) holds for $\mathbf{X}(t)$. Now the probability distribution of $\mathbf{Z}_\varepsilon(t)$ has the density (at $\mathbf{x}$) $\varepsilon^{-n} p(\varepsilon^{-2}t; \mathbf{z}, \varepsilon^{-1}\mathbf{x} + \varepsilon^{-2} t U_0 \bar{\mathbf{b}})$ if $\mathbf{X}(0) = \mathbf{z}$. Hence the CLT asserts that $\varepsilon^{-n} p(\varepsilon^{-2}t; \mathbf{z}, \varepsilon^{-1}\mathbf{x} + \varepsilon^{-2} t U_0 \bar{\mathbf{b}})\, d\mathbf{x}$ converges weakly, as $\varepsilon \downarrow 0$, to the Gaussian distribution:

$$\phi(t, \mathbf{x})\, d\mathbf{x} = (2\pi t)^{-n/2} (\operatorname{Det} \mathbf{K})^{-1/2} \exp\left\{ -\frac{1}{2t} \sum_{i,j=1}^{n} K^{ij} x_i x_j \right\} d\mathbf{x}. \tag{1.5}$$

Here K^{ij} is the (i, j) element of the matrix $\mathbf{K}^{-1}$. Thus as $\varepsilon \downarrow 0$ we obtain

$$\varepsilon^{-n} C(\varepsilon^{-1}\mathbf{x} + \varepsilon^{-2} t U_0 \bar{\mathbf{b}}, \varepsilon^{-2} t)\, d\mathbf{x} \to C_0 \phi(t, \mathbf{x})\, d\mathbf{x} \tag{1.6}$$

where C_0 is the total initial concentration.

From here on we will refer to $((D_{ij}))$ as the *small scale dispersion matrix* and $((K_{ij}))$ as the *large scale dispersion matrix.*

CLTs such as described above have been derived for periodic coefficients D_{ij}, b_i in Bensoussan, Lions, and Papanicolaou (1978) and Bhattacharya (1985). Under the assumption that the elliptic operator on the right-hand side of (1.1) is self-adjoint, Kozlov (1979), (1980), and Papanicolaou and Varadhan (1979) have proved such CLTs for the case where the coefficients are stationary, ergodic random fields. An extension to the nonself-adjoint case for almost periodic coefficients, when the large scale velocity $\bar{\mathbf{b}}$ is nonzero, is given in Bhattacharya and Ramasubramanian (1988). Papanicolaou and Pironeau (1981) also deal with a nonself-adjoint case when the coefficients constitute a general ergodic random field and $\bar{\mathbf{b}} = 0$.

Such problems arise in analyzing the movement of contaminants in saturated porous media such as aquifers as well as in laboratory columns. The dependence of $\mathbf{K}$ on U_0 has been studied experimentally in laboratory columns (see, e.g., Fried and Combarnous (1971)). The spatial scale parameter a is fixed in such experiments. In aquifers, on the other hand, the main interest from the point of view of long term prediction lies in the analysis of $\mathbf{K}$ as a function of the scale parameter a for a fixed velocity field, and therefore for a fixed U_0 (Gupta and Bhattacharya, (1986)). Field scale dispersions in aquifers have been analyzed for the ergodic random field case (when $\bar{b}$ is nonzero) in, e.g., Gelhar and Axness (1983), Winter, Newman, and Neuman (1984), and Dagan (1984). For certain classes of periodic coefficients, the dependence of $\mathbf{K}$ on a and U_0 has been analyzed in Gupta and Bhattacharya (1986) and Guven and Molz (1986). A more detailed survey of the hydrologic literature is given in Sposito, Jury, and Gupta (1986).

The dependence of $\mathbf{K}$ on U_0 has been treated in the literature separately from its dependence on a because of the physical contexts in which these arise. As we shall see in § 2, the roles of U_0 and a in this respect are interchangeable. Indeed $\mathbf{K}$ depends only on the product aU_0.

In § 3 we analyze the dependence of $\mathbf{K}$ on aU_0 for the class of periodic coefficients such that D_{ij}'s are constants and $\mathbf{b}$ has zero divergence. It is shown that for one broad class of periodic coefficients, the K_{ii}'s grow quadratically as $aU_0 \to \infty$, and that the K_{ii}'s approach asymptotic constancy for another class.

Section 4 provides a refinement of the Gaussian approximation (1.6) in the form of an asymptotic expansion in powers of ε. In probability theory such an expansion is called a *Cramer-Edgeworth expansion.* In the differential equations literature it is referred to as a *singular perturbation expansion.* For prediction of concentration $C(\mathbf{x}, t)$ in aquifers over time scales that are not very large, such expansions provide better approximations than the Gaussian. The importance of predictions over such time scales has been discussed, for example, by Guven and Molz (1986) and Dagan (1984).

2. Interchangeability of velocity and spatial scale parameters in K. Write,

$$\mathbf{K}(U_0, a) = \mathbf{K}, \qquad K_{ij}(U_0, a) = K_{ij}, \tag{2.1}$$

indicating the dependence of the large scale dispersion matrix $\mathbf{K}$ on the velocity and scale parameters U_0 and a.

PROPOSITION 2.1. *If the central limit theorem holds for the solution* $\mathbf{X}(t)$ *of* (1.3), *then* $\mathbf{K}$ *depends on* U_0 *and* a *only through their product* aU_0. *In particular,*

$$\mathbf{K}(U_0, a) = \mathbf{K}(a, U_0) = \mathbf{K}(aU_0, 1). \tag{2.2}$$

To prove this, express the solution of (1.3) as $\mathbf{X}(t; a, U_0)$ to indicate its dependence on a and U_0. Define the stochastic process

$$\mathbf{Y}(t; a, U_0) = a\mathbf{X}(t/a^2; 1, U_0). \tag{2.3}$$

Then $\mathbf{Y}(t; a, U_0)$ satisfies the Itô equation

$$\begin{aligned} d\mathbf{Y}(t; a, U_0) &= aU_0\mathbf{b}(\mathbf{X}(t/a^2; 1, U_0))\frac{dt}{a^2} + a\sigma(\mathbf{X}(t/a^2; 1, U_0))\, d\mathbf{B}(t/a^2) \\ &= \frac{U_0}{a}\mathbf{b}(\mathbf{Y}(t; a, U_0)/a)\, dt + \sigma(\mathbf{Y}(t; a, U_0)/a)\, d\bar{\mathbf{B}}(t), \end{aligned} \tag{2.4}$$

where $\bar{\mathbf{B}}(t)$ is defined by

$$d\bar{\mathbf{B}}(t) = a\, d\mathbf{B}(t/a^2), \qquad \bar{\mathbf{B}}(0) = \mathbf{B}(0) = 0. \tag{2.5}$$

Note that $\bar{\mathbf{B}}(t)$ is, like $\mathbf{B}(t)$, a standard n-dimensional Brownian motion. It now follows from (2.4) that $\mathbf{Y}(t; a, U_0)$ has the same distribution as $\mathbf{X}(t; a, U_0/a)$ (with the initial value $\mathbf{Y}(0; a, U_0) = a\mathbf{z}$). Hence

$$\lim_{t\to\infty} \frac{\operatorname{Var} \mathbf{Y}(t; a, U_0)}{t} = \lim_{t\to\infty} \frac{\operatorname{Var} \mathbf{X}(t; a, U_0/a)}{t} = \mathbf{K}(U_0/a, a), \tag{2.6}$$

where Var stands for the variance-covariance matrix. Now, from (2.3),

$$\begin{aligned} \lim_{t\to\infty} \frac{\operatorname{Var} \mathbf{Y}(t; a, U_0)}{t} &= \lim_{t\to\infty} a^2 \frac{\operatorname{Var} \mathbf{X}(t/a^2; 1, U_0)}{t} \\ &= \lim_{t\to\infty} \frac{\operatorname{Var} \mathbf{X}(t/a^2; 1, U_0)}{t/a^2} = \mathbf{K}(U_0, 1). \end{aligned} \tag{2.7}$$

Relations (2.6) and (2.7) yield,

$$\mathbf{K}(U_0/a, a) = \mathbf{K}(U_0, 1). \tag{2.8}$$

Write $\alpha = U_0/a$, $\beta = a$. Then (2.8) becomes

$$\mathbf{K}(\alpha, \beta) = \mathbf{K}(\alpha\beta, 1) \quad \text{for all } \alpha > 0, \quad \beta > 0. \tag{2.9}$$

This proves the proposition.

It may be remarked that in a periodic model a is simply the period (Gupta and Bhattacharya (1986)). In an ergodic random field model (see Gelhar and Axness (1983), Winter et al. (1984)), a may be taken to be the characteristic correlation length. Fried and Combarnous (1971) give an account of the fairly extensive laboratory experiments that have been done to study the effect of increase in velocity on dispersion in porous media. A broad mathematical justification of these experimentally observed relationships appears in Bhattacharya and Gupta (1983). In these studies the spatial scale is held fixed at the so-called *Darcy level*, while velocity is increased. On the other hand, dependence of dispersion on large spatial scales has been analyzed in field situations for various models of heterogeneous porous media. The above proposition shows that the two relationships are mathematically equivalent. For this reason, in the next section the spatial scale a is held fixed at $a = 1$, while the velocity parameter U_0 is allowed to vary.

3. An expansion of the large scale dispersion in the periodic model. In (1.1), take D_{ij}'s to be constants and b_i's continuously differentiable periodic functions satisfying the divergence condition

$$\operatorname{div} \mathbf{b} = 0. \tag{3.1}$$

In view of proposition (2.1), we take the period of b_i to be one in each coordinate without loss of generality. Let L denote the elliptic operator

$$Lg(\mathbf{x}) = Dg(\mathbf{x}) + U_0 \mathbf{b}(\mathbf{x}) \cdot \nabla g(\mathbf{x}), \qquad \mathbf{x} \in \mathfrak{R}^n \tag{3.2}$$

where

$$D = \frac{1}{2} \sum D_{ij} \frac{\partial^2}{\partial x_i \, \partial x_j}. \tag{3.3}$$

Let $T = [0, 1]^n$. Define

$$\bar{b}_i = \int_T b_i(\mathbf{x}) \, d\mathbf{x}, \qquad i = 1, 2, \cdots, n, \tag{3.4}$$

and let g_i be a periodic function satisfying

$$Lg_i = b_i - \bar{b}_i. \tag{3.5}$$

Then it follows from Bhattacharya (1985) that the large scale dispersion coefficients are given by

$$K_{ij} = D_{ij} - U_0^2 \int_T g_i(\mathbf{x})(b_j(\mathbf{x}) - \bar{b}_j) \, d\mathbf{x} - U_0^2 \int_T g_j(\mathbf{x})(b_i(\mathbf{x}) - \bar{b}_i) \, d\mathbf{x}. \tag{3.6}$$

It is convenient to work with the following spaces of (equivalence classes of) complex-valued functions on T:

$$H^0 = \left\{ h \colon \int_T |h(\mathbf{x})|^2 \, d\mathbf{x} < \infty, \int_T h(\mathbf{x}) \, d\mathbf{x} = 0, \right.$$
$$\left. \text{and } h \text{ satisfies periodic boundary conditions} \right\},$$

$$H^1 = \left\{ h \in H^0 \colon \int_T |\nabla h(\mathbf{x})|^2 \, d\mathbf{x} < \infty \right\},$$

$$H^2=\left\{h\in H^1: \int_T \sum_{i,j=1}^n \left|\frac{\partial^2}{\partial x_i\,\partial x_j}h(\mathbf{x})\right|^2 d\mathbf{x}<\infty\right\}.$$

Here, $|\cdot|$ denotes both absolute value and Euclidean norm. For convenience, we take the norm and inner product on H^1 to be

$$\|h\|_1^2=\int_T \sum_{i,j=1}^n D_{ij}\frac{\partial}{\partial x_i}h(\mathbf{x})\frac{\partial}{\partial x_j}\bar{h}(\mathbf{x})\,d\mathbf{x}, \quad \text{and}$$

$$\langle h, w\rangle_1=\int_T \sum_{i,j=1}^n D_{ij}\frac{\partial}{\partial x_i}h(\mathbf{x})\frac{\partial}{\partial x_j}\bar{w}(\mathbf{x})\,d\mathbf{x}$$

for $h, w\in H^1$. This is allowed, since $((D_{ij}))$ is a real, positive-definite, symmetric matrix and

$$\int_T h(\mathbf{x})\,d\mathbf{x}=0$$

for $h\in H^1$.

For a given set f_i $(i=1,2,\cdots,n)$ in H^1 let g_i be the solutions in H^2 of

$$Lg_i=f_i. \tag{3.7}$$

Standard results in the theory of elliptic partial differential operators imply that (3.7) has a unique solution $g_i\in H^2$ for each $f_i\in H^1$.

Throughout we shall write

$$E_{ij}=E_{ij}(U_0)=-U_0^2\int_T g_i(\mathbf{x})f_j(\mathbf{x})\,d\mathbf{x}. \tag{3.8}$$

In this notation, $K_{ij}=D_{ij}+E_{ij}+E_{ji}$ with $f_i=b_i-\bar{b}_i$.

Note that the operator D is one to one on H^2 onto H^0. To obtain useful eigenfunction expansions we note that for $f\in H^0$ and $g\in H^2$, $Lg=f$ if and only if $[I+U_0H]g=D^{-1}f$, where $Hg(\mathbf{x})=D^{-1}\mathbf{b}(\mathbf{x})\cdot\nabla g(\mathbf{x})$. We can consider H as an operator from H^1 to itself; as such, it is compact and skew-symmetric. Then H has eigenfunctions $\{\phi_k\}_{k=1,2,\cdots}$ and corresponding eigenvalues $\{\sqrt{-1}\,\lambda_k\}_{k=1,2,\cdots}$ with the following properties:

(i) Each λ_k is real and $\lim_{k\to\infty}\lambda_k=0$.

(ii) $\{\phi_k\}_{k=1,2,\cdots}$ is a complete orthonormal set on $H^1\cap N^\perp$, where $N=\{h\in H^1: Hh=0\}$ is the null space of H in H^1 and $\perp$ denotes orthogonal complement.

(iii) Each $h\in H^1$ can be represented as

$$h=h_N+\sum_{k=1}^\infty \alpha_k\phi_k, \tag{3.9}$$

where $h_N\in N$ and for $k=1,2,\cdots$, $\alpha_k=\langle h,\phi_k\rangle_1$. Note that

$$\|h\|_1^2=\|h_N\|_1^2+\sum_{k=1}^\infty|\alpha_k|^2, \quad \text{and}$$

$$Hh=\sum_{k=1}^\infty \sqrt{-1}\,\lambda_k\alpha_k\phi_k.$$

Suppose that for $g\in H^2$ and $f\in H^0$, the representation (3.9) becomes

$$g=g_N+\sum_{k=1}^\infty \alpha_k\phi_k, \quad \text{and}$$

$$D^{-1}f=(D^{-1}f)_N+\sum_{k=1}^\infty \beta_k\phi_k.$$

Then $Lg=f$ if and only if $[I+U_0H]g=D^{-1}f$, i.e.,

$$g_N+\sum_{k=1}^{\infty}(1+\sqrt{-1}\,U_0\lambda_k)\alpha_k\phi_k=(D^{-1}f)_N+\sum_{k=1}^{\infty}\beta_k\phi_k,$$

i.e.,

$$g=L^{-1}f=(D^{-1}f)_N+\sum_{k=1}^{\infty}\frac{\beta_k}{1+\sqrt{-1}\,U_0\lambda_k}\phi_k. \tag{3.10}$$

Suppose that the given set of functions f_i is real valued and contained in H^0. If we have

$$D^{-1}f_i=(D^{-1}f_i)_N+\sum_{k=1}^{\infty}\beta_{ik}\phi_k$$

for each i, then (3.10) gives

$$g_i=L^{-1}f_i=(D^{-1}f_i)_N+\sum_{k=1}^{\infty}\frac{\beta_{ik}}{1+\sqrt{-1}\,U_0\lambda_k}\phi_k.$$

It follows that for general i and j

$$\begin{aligned}E_{ij}(U_0)&=-U_0^2\int_T g_i(\mathbf{x})DD^{-1}f_j(\mathbf{x})\,d\mathbf{x}=U_0^2\langle g_i,D^{-1}f_j\rangle_1\\&=U_0^2\left\{\langle(D^{-1}f_i)_N,(D^{-1}f_j)_N\rangle_1+\sum_{k=1}^{\infty}\frac{\beta_{ik}\bar{\beta}_{jk}}{1+\sqrt{-1}\,U_0\lambda_k}\right\}.\end{aligned} \tag{3.11}$$

If $i=j$, a sharper result can be obtained. We have

$$\begin{aligned}E_{ii}(U_0)&=-U_0^2\int_T g_i(\mathbf{x})D[I+U_0H]g_i(\mathbf{x})\,d\mathbf{x}\\&=U_0^2\{\|g_i\|_1^2+U_0\langle g_i,Hg_i\rangle_1\}.\end{aligned}$$

Since H is skewsymmetric on H^1 and g_i is real-valued, $\langle g_i,Hg_i\rangle_1=0$. Consequently,

$$E_{ii}(U_0)=U_0^2\left\{\|(D^{-1}f_i)_N\|_1^2+\sum_{k=1}^{\infty}\frac{|\beta_{ik}|^2}{1+U_0^2\lambda_k^2}\right\}. \tag{3.12}$$

It may not be apparent how to obtain (3.12) by taking $j=i$ in (3.11). The two formulas can be reconciled by noting the following:

(i) Since $((D_{ij}))$ and b_i are real, for each eigenfunction-eigenvalue pair ϕ_k, $\sqrt{-1}\,\lambda_k$ there is a complex conjugate pair $\phi_l=\bar{\phi}_k$, $\sqrt{-1}\,\lambda_l=-\sqrt{-1}\,\lambda_k$.

(ii) For such conjugate pairs,

$$\beta_{ik}=\langle f_i,\phi_k\rangle_1=\langle f_i,\bar{\phi}_l\rangle_1=\bar{\beta}_{il}$$

since f_i is real.

(iii) Then for such pairs,

$$\frac{|\beta_{ik}|^2}{1+\sqrt{-1}\,U_0\lambda_k}+\frac{|\beta_{il}|^2}{1+\sqrt{-1}\,U_0\lambda_l}=\frac{|\beta_{ik}|^2}{1+U_0^2\lambda_k^2}+\frac{|\beta_{il}|^2}{1+U_0^2\lambda_l^2}.$$

3.1. Applications and examples. Expressions (3.11) and (3.12) are our basic tools for analyzing the behavior of the E_{ij}'s and K_{ij}'s. In the following, we show how these expressions can be applied to the examples of Gupta and Bhattacharya (1986) as well as to new examples, and we give some results that illustrate how they can be used to obtain general statements.

It is obvious from (3.11) and (3.12) that $E_{ij}(U_0)=O(U_0^2)$ if $\langle(D^{-1}f_i)_N,(D^{-1}f_j)_N\rangle_1\neq 0$ and $E_{ij}(U_0)=o(U_0^2)$ otherwise. In particular, $E_{ii}(U_0)=O(U_0^2)$ if $(D^{-1}f_i)_N\neq 0$ and $E_{ii}(U_0)=o(U_0^2)$ otherwise. We note that $N=\{h\in H^1\colon Hh=0\}$ is just the null space of $\mathbf{b}\cdot\nabla$ in H^1, i.e., the set of $h\in H^1$ such that $\mathbf{b}(\mathbf{x})\cdot\nabla h(\mathbf{x})=0$ almost everywhere in T. This is to say that N is the set of elements of H^1 that are constant along the *flow curves* determined by $\mathbf{b}$. By a flow curve, we mean a characteristic of the partial differential operator $\mathbf{b}\cdot\nabla$, i.e., a solution of the autonomous system $\dot{\mathbf{x}}=\mathbf{b}(\mathbf{x})$.

LEMMA 3.1. *Suppose that $f_i\in H^1$ is constant along each flow curve. Then either $f_i=0$, in which case $E_{ij}(U_0)=0$ for each j, or $E_{ii}(U_0)=O(U_0^2)$.*

Proof. We have that $f_i\in N$ and

$$\langle f_i,D^{-1}f_i\rangle_1=-\int_T f_i(\mathbf{x})^2\,d\mathbf{x}.$$

It follows that if $f_i\neq 0$, then $(D^{-1}f_i)_N\neq 0$ and $E_{ii}(U_0)=O(U_0^2)$.

Example 3.2 (Gupta and Bhattacharya (1986)). Take $n=3$, and,

$$\mathbf{b}(\mathbf{x})=(1+\sin 2\pi x_3,\sin 2\pi x_3,0),\qquad \mathbf{x}=(x_1,x_2,x_3).$$

Then it is simple to check that for $i=1,2$, $f_i=b_i-\bar{b}_i$ satisfies the hypothesis of Lemma 3.1 and E_{ii} and K_{ii} are $O(U_0^2)$. In fact, f_1 and f_2 depend only on x_3, and so $D^{-1}f_1$ and $D^{-1}f_2$ depend only on x_3. Then $D^{-1}f_1$ and $D^{-1}f_2$ are in N, and since

$$\langle D^{-1}f_1,D^{-1}f_2\rangle_1=-\int_T \sin 2\pi x_3 D^{-1}\sin 2\pi x_3\,dx>0,$$

it follows that E_{12}, E_{21}, and K_{12} are $O(U_0^2)$.

As an operator on H^1, $\mathbf{b}\cdot\nabla$ has range

$$R=\{f\in H^0\colon f=\mathbf{b}\cdot\nabla h \text{ for some } h\in H^1\}$$

in H^0. This range R, as well as the null space N, can be helpful in determining the behavior of the E_{ij}'s and K_{ij}'s.

LEMMA 3.3. *Suppose that $f_i\in R$. Then*

$$\lim_{U_0\to\infty} E_{ii}(U_0)=\|h_i\|_1^2,$$

where h_i is the unique element of $H^1\cap N^\perp$ such that $f_i=\mathbf{b}\cdot\nabla h_i$. Also for $i\neq j$,

$$E_{ij}(U_0)=O(U_0)\simeq U_0\langle h_i,D^{-1}f_j\rangle_1,\quad \textit{and} \tag{3.13}$$

$$E_{ji}(U_0)=O(U_0)\simeq -U_0\langle D^{-1}f_j,h_i\rangle_1, \tag{3.14}$$

for large U_0.

Remark. In (3.13) and (3.14), $\simeq$ means that after division by U_0, both sides approach the same limit as U_0 approaches infinity. In particular, if the inner products in (3.13) and (3.14) are zero, then $E_{ij}(U_0)$ and $E_{ji}(U_0)$ are $o(U_0)$.

Proof. It is clear that h_i exists, and we write

$$h_i=\sum_{k=1}^{\infty}\gamma_{ik}\phi_k.$$

Then

$$D^{-1}f_i=Hh_i=\sum_{k=1}^{\infty}\sqrt{-1}\,\lambda_k\gamma_{ik}\phi_k,$$

and (3.12) gives

$$E_{ii}(U_0)=U_0^2\sum_{k=1}^{\infty}\frac{\lambda_k^2|\gamma_{ik}|^2}{1+U_0^2\lambda_k^2}.$$

Thus

$$\lim_{U_0\to\infty}E_{ii}(U_0)=\sum_{k=1}^{\infty}|\gamma_{ik}|^2=\|h_i\|_1^2.$$

For $j\neq i$, we write

$$D^{-1}f_j=(D^{-1}f_j)_N+\sum_{k=1}^{\infty}\beta_{jk}\phi_k,$$

and (3.11) gives

$$E_{ij}(U_0)=U_0\sum_{k=1}^{\infty}\frac{\sqrt{-1}\,U_0\lambda_k\gamma_{ik}\bar{\beta}_{jk}}{1+\sqrt{-1}\,U_0\lambda_k}. \tag{3.15}$$

Then for large U_0,

$$E_{ij}(U_0)=O(U_0)\simeq U_0\sum_{k=1}^{\infty}\gamma_{ik}\bar{\beta}_{jk}=U_0\langle h_i,D^{-1}f_j\rangle_1.$$

Similarly,

$$\begin{aligned}E_{ji}&=-U_0\sum_{k=1}^{\infty}\frac{\sqrt{-1}\,U_0\lambda_k\beta_{jk}\bar{\gamma}_{ik}}{1+\sqrt{-1}\,U_0\lambda_k}=O(U_0)\\&\simeq -U_0\sum_{k=1}^{\infty}\beta_{jk}\bar{\gamma}_{ik}=-U_0\langle D^{-1}f_j,h_i\rangle_1\end{aligned} \tag{3.16}$$

for large U_0.

It is interesting to note the behavior of K_{ij} when $f_i\equiv b_i-\bar{b}_i$ belongs to R. From (3.15), (3.16), and an extension of the reasoning after (3.12), we obtain

$$E_{ij}(U_0)+E_{ji}(U_0)=2U_0\sum_{k=1}^{\infty}\frac{\sqrt{-1}\,U_0\lambda_k}{1+U_0^2\lambda_k^2}\gamma_{ik}\bar{\beta}_{jk}. \tag{3.17}$$

Since the sum on the right-hand side of (3.17) approaches zero as U_0 grows large, K_{ij} is $o(U_0)$ for large U_0 when $f_i\in R$. More can be said if f_j as well as f_i is in R. Suppose $f_j\in R$ and

$$h_j=\sum_{k=1}^{\infty}\gamma_{jk}\phi_k$$

is the unique element of $H^1\cap N^\perp$ such that $f_j=\mathbf{b}\cdot\nabla h_j$. Taking $\beta_{jk}=\sqrt{-1}\,\lambda_k\gamma_{jk}$ in (3.17) gives

$$E_{ij}(U_0)+E_{ji}(U_0)=2\sum_{k=1}^{\infty}\frac{U_0^2\lambda_k^2}{1+U_0^2\lambda_k^2}\gamma_{ik}\bar{\gamma}_{jk},$$

and so

$$\begin{aligned}\lim_{U_0\to\infty}K_{ij}(U_0)&=D_{ij}-\lim_{U_0\to\infty}\{E_{ij}(U_0)+E_{ji}(U_0)\}\\&=D_{ij}-2\langle h_i,h_j\rangle_1.\end{aligned}$$

Unfortunately, we cannot characterize the range R without making restrictive assumptions about $\mathbf{b}$. We can imagine many applications in which one of the b_i's never vanishes on T, and so to be specific we assume for the remainder of this section that $b_1 > 0$ on T. This allows us to parameterize the flow curves in terms of x_1. Indeed, if we write $\mathbf{x} \in \mathfrak{R}^n$ as $\mathbf{x} = (x_1, \hat{\mathbf{x}})$ for $\hat{\mathbf{x}} = (x_2, \cdots, x_n) \in \mathfrak{R}^{n-1}$, then the flow curves are just the curves $(t, \hat{\mathbf{x}}(t))$, where $\hat{\mathbf{x}}(t)$ solves the nonautonomous system

$$\hat{\mathbf{x}}' = \hat{\mathbf{b}}(t, \hat{\mathbf{x}}) = \left(\frac{b_2(t, \hat{\mathbf{x}})}{b_1(t, \hat{\mathbf{x}})}, \cdots, \frac{b_n(t, \hat{\mathbf{x}})}{b_1(t, \hat{\mathbf{x}})}\right).$$

In fact, for each value of $\hat{\mathbf{x}}(0) \in \mathfrak{R}^{n-1}$, this system determines a unique curve $(t, \hat{\mathbf{x}}(t))$ in the strip $S = [0, 1] \times \mathfrak{R}^{n-1}$, which is defined for $0 \leqq t \leqq 1$; furthermore, each $\mathbf{x} \in T$ can be uniquely written as $\mathbf{x} = (x_1, \hat{\mathbf{x}}(x_1)$, a point on such a curve. (The periodicity assumption on $\mathbf{b}$ implies that $\hat{\mathbf{b}}$ is defined and bounded everywhere.) We identify functions on T satisfying periodic boundary conditions with periodic functions on S in the obvious way.

LEMMA 3.4. *Suppose $f \in C^1$ is a function on T that satisfies*

$$\int_0^1 \frac{f(t, \hat{\mathbf{x}}(t))}{b_1(t, \hat{\mathbf{x}}(t))} \, dt = 0, \tag{3.18}$$

for every flow curve $(t, \hat{\mathbf{x}}(t))$, $0 \leqq t \leqq 1$. Then $f \in R$.

Proof. For each $\mathbf{x} \in T$, we write uniquely $\mathbf{x} = (x_1, \hat{\mathbf{x}}(x_1))$ for a flow curve $(t, \hat{\mathbf{x}}(t))$ and define

$$h(\mathbf{x}) = \int_0^{x_1} \frac{f(t, \hat{\mathbf{x}}(t))}{b_1(t, \hat{\mathbf{x}}(t))} \, dt.$$

Since f and b_1 are C^1, so is h. Furthermore, for $\mathbf{x} \in T$,

$$\mathbf{b}(\mathbf{x}) \cdot \nabla h(\mathbf{x}) = b_1(\mathbf{x}) \frac{d}{dx_1} h(x_1, \hat{\mathbf{x}}(x_1)) = f(\mathbf{x}).$$

Clearly, $h(0, \hat{\mathbf{x}}) = 0$ for all $\hat{\mathbf{x}}$ and $h(x_1, \hat{\mathbf{x}})$ satisfies periodic boundary conditions in $\hat{\mathbf{x}}$ for $0 < x_1 < 1$. Also, (3.18) implies that $h(1, \hat{\mathbf{x}}) = 0$ for all $\hat{\mathbf{x}}$. Then $h \in H^1$ and $f \in R$.

COROLLARY 3.5. *Suppose that $f_i \in C^1$ and satisfies*

$$\int_0^1 \frac{f_i(t, \hat{\mathbf{x}}(t))}{b_1(t, \hat{\mathbf{x}}(t))} \, dt = 0$$

for every flow curve $(t, \hat{\mathbf{x}}(t))$, $0 \leqq t \leqq 1$. Then the conclusions of Lemma 3.3 *hold.*

Example 3.6 (Gupta and Bhattacharya (1986)). Take $n = 3$, and

$$\mathbf{b}(\mathbf{x}) = (\bar{b}_1, 1 + \sin 2\pi x_1, \sin 2\pi x_1).$$

Then $f_i = b_i - \bar{b}_i$, $i = 1, 2, 3$, satisfy the hypothesis of Lemma 3.4, and each E_{ii} and K_{ii} is $O(1)$. It follows from the remarks after the proof of Lemma 3.3 that each K_{ij} is $O(1)$.

In Example 3.2, each E_{ij} and K_{ij} is $O(U_0^2)$ for $i, j = 1, 2$; in Example 3.6, each E_{ij} and K_{ij} is $O(1)$. We give an additional example in which E_{22} and K_{22} are $O(U_0^2)$ and all other E_{ij}'s and K_{ij}'s are $O(1)$.

Example 3.7. Let $n = 3$, and $b_3(\mathbf{x}) = 2 + (\cos 2\pi x_1)(\cos 2\pi x_2)$, $b_1(x) = 2 + \sin 2\pi x_1$, $b_1(\mathbf{x}) = 0$. Then $E_{11} = 0$ and $K_{11} = D_{11}$. Also, clearly, $E_{12} = E_{13} = E_{21} = E_{31} = 0$ and $K_{13} = D_{13}$, $K_{12} = D_{12}$. Since $\mathbf{b} \cdot \nabla b_2 = 0$, E_{22} and K_{22} are $O(U_0^2)$ by Lemma 3.1. Now the coefficients of L do not involve x_3. Hence, the solution of $Lg_3(\mathbf{x}) = b_3(\mathbf{x}) - \bar{b}_3$ is of the form $g_3(\mathbf{x}) = g(x_1, x_2)$ where

$$\frac{1}{2} \sum_{i,j=1}^{2} D_{ij} \frac{\partial^2 g}{\partial x_i \, \partial x_j} + U_0(2 + \sin 2\pi x_1) \frac{\partial g}{\partial x_2} = (\cos 2\pi x_2)(\cos 2\pi x_1). \tag{3.19}$$

Since $b_2 > 0$, it follows from Lemma 3.4 (with $n = 2$) applied to the function f on the right-hand side of (3.19) that E_{33} is $O(1)$ and $K_{33} = D_{33} + O(1)$. A direct computation shows that $E_{23} = E_{32} = 0$ and $K_{23} = D_{23}$.

The above examples subtly reflect the influence of the geometry of the flow curves on the asymptotic behavior of the E_{ij}'s and K_{ij}'s. The tools developed here can be used to bring out this geometrical influence. In the remainder of this section, we illustrate how this can be done by making assumptions about the geometry of the flow curves and obtaining statements about the asymptotic behavior of the E_{ij}'s and K_{ij}'s. While these statements apply only to somewhat specialized situations, they and their proofs suggest promising directions for future work. They also show the type of asymptotic behavior that is possible in situations that come naturally to mind. Our first result is another corollary of Lemma 3.4.

COROLLARY 3.8. *Suppose that for some i,* $2 \leqq i \leqq n$, *every flow curve is periodic in the ith component, i.e.,* $x_i(0) = x_i(1)$ *for every flow curve* $(t, \hat{\mathbf{x}}(t)) = (t, x_2(t), \cdots, x_n(t))$, $0 \leqq t \leqq 1$. *Then* $\bar{b}_i = 0$ *and the conclusions of Lemma* 3.3 *hold.*

Proof. We have

$$0 = x_i(1) - x_i(0) = \int_0^1 x_i'(t)\, dt = \int_0^1 b_i(t, \hat{\mathbf{x}}(t))/b_1(t, \hat{\mathbf{x}}(t))\, dt$$

for every flow curve $(t, \hat{\mathbf{x}}(t))$, $0 \leqq t \leqq 1$. It follows from Lemma 3.4 that $b_i \in R$, i.e., $b_i = \mathbf{b} \cdot \nabla h$ for some $h \in H^1$. Then

$$\bar{b}_i = \int_T \mathbf{b}(\mathbf{x}) \cdot \nabla h(\mathbf{x})\, d\mathbf{x} = 0,$$

which implies $f_i = b_i - \bar{b}_i = b_i \in R$.

The examples given previously have the property that each E_{ij} and K_{ij} is either $O(U_0^2)$ or $O(1)$. An important unresolved question is whether any other behavior is possible in general. We show now that under an additional restriction on the flow curves, i.e., on $\mathbf{b}$, each E_{ii} and K_{ii} must be either $O(U_0^2)$ or $O(1)$.

We assume not only that $b_1 > 0$ in T but also that the difference between any two flow curves is constant as x_1 varies. This is equivalent to assuming that for $i = 2, \cdots, n$, the ratio $b_i(\mathbf{x})/b_1(\mathbf{x})$ depends only on x_1. Under this assumption, the flow curves can be conveniently described as follows: Let $(t, \hat{\mathbf{x}}(t))$, $0 \leqq t \leqq 1$, be the flow curve passing through the origin, i.e., such that $\hat{\mathbf{x}}(0) = 0$; then every other flow curve can be written as $(t, \hat{\mathbf{x}}_0 + \hat{\mathbf{x}}(t))$, $0 \leqq t \leqq 1$, for an appropriate $\hat{\mathbf{x}}_0$.

PROPOSITION 3.9. *Under the present assumptions,* b_1 *is constant along each flow curve and either* $b_1 \equiv \bar{b}_1$, *in which case* $E_{11}(U_0) = 0$, *or* $E_{11}(U_0) = O(U_0^2)$.

Proof. We have that

$$\hat{\mathbf{x}}'(x_1) = (b_2(\mathbf{x})/b_1(\mathbf{x}), \cdots, b_n(\mathbf{x})/b_1(\mathbf{x})),$$

and so $\mathbf{b}(\mathbf{x}) = b_1(\mathbf{x})(1, \hat{\mathbf{x}}'(x_1))$. Then the assumption that $\nabla \cdot \mathbf{b}(\mathbf{x}) \equiv 0$ implies

$$(1, \hat{\mathbf{x}}'(x_1)) \cdot \nabla b_1(\mathbf{x}) = 0.$$

But this is to say that the directional derivative of b_1 along each flow curve is zero, and the proposition follows from Lemma 3.1.

THEOREM 3.10. *Under the present assumptions, either* $E_{ii}(U_0) = O(U_0^2)$ *or* $f_i \in R$ *and the conclusions of Lemma* 3.3 *hold.*

Proof. If $(D^{-1}f_i)_N \neq 0$, then $E_{ii}(U_0) = O(U_0^2)$. Suppose $(D^{-1}f_i)_N = 0$, i.e., that $\langle h, D^{-1}f_i \rangle_1 = 0$ for every $h \in N$. We show that $f_i \in R$.

Set $\hat{T}=\{\hat{\mathbf{x}}=(x_2,\cdots,x_n)\in\mathfrak{R}^{n-1}\colon -\frac{1}{2}\leqq x_i\leqq\frac{1}{2}, 2\leqq i\leqq n\}$, and denote by $\hat{\delta}$ the restriction of the Dirac delta distribution on $\mathfrak{R}^{n-1}$ to $\hat{T}$. Let $\{\hat{\psi}_k\}_{k=1,2,\cdots}$ be a sequence of C^∞ functions on $\hat{T}$ such that each $\hat{\psi}_k$ has support in the interior of $\hat{T}$ and

$$\lim_{k\to\infty}\hat{\psi}_k=\hat{\delta}$$

in the distributional sense. Extend $\hat{\delta}$ and each $\hat{\psi}_k$ to be periodic with period one in each variable over all $\mathfrak{R}^{n-1}$.

Let $(t,\hat{\mathbf{x}}_0+\hat{\mathbf{x}}(t))$, $0\leqq t\leqq 1$, be an arbitrary flow curve. For $\mathbf{x}=(x_1,\hat{\mathbf{x}})\in S$, define

$$\psi_k(\mathbf{x})=\hat{\psi}_k(\hat{\mathbf{x}}-\hat{\mathbf{x}}_0-\hat{\mathbf{x}}(x_1)),\qquad k=1,2,\cdots.$$

Each ψ_k is constant along every flow curve and so belongs to N. Also, for $\mathbf{x}=(x_1,\hat{\mathbf{x}})$,

$$\lim_{k\to\infty}\psi_k(\mathbf{x})=\hat{\delta}(\hat{\mathbf{x}}-\hat{\mathbf{x}}_0-\hat{\mathbf{x}}(x_1)) \tag{3.20}$$

in the sense of distributions on $\mathfrak{R}^{n-1}$. Then

$$\begin{aligned} 0&=\lim_{k\to\infty}\langle\psi_k, D^{-1}f_i\rangle_1\\ &=\lim_{k\to\infty}-\int_T\psi_k(\mathbf{x})f_i(\mathbf{x})\,d\mathbf{x} \qquad (3.21)\\ &=-\int_0^1 f_i(t,\hat{\mathbf{x}}_0+\hat{\mathbf{x}}(t))\,dt.\end{aligned}$$

The last equality follows from (3.20) by periodicity even when the flow curve is not contained in T. Since b_1 is constant along the flow curve by Proposition 3.9, (3.21) implies

$$\int_0^1\frac{f_i(t,\hat{\mathbf{x}}_0+\hat{\mathbf{x}}(t))}{b_1(t,\hat{\mathbf{x}}_0+\hat{\mathbf{x}}(t))}\,dt=0.$$

Since the flow curve is arbitrary, it follows from Corollary 3.5 that $f_i\in R$.

We offer a final example on which Corollary 3.8, Proposition 3.9, and Theorem 3.10 are applicable.

Example 3.11. Let ξ be any C^2 function on $\mathfrak{R}^1$ that is periodic with period one and such that $\xi(0)=\xi(1)=0$. We take $n=2$ and construct $\mathbf{b}\colon\mathfrak{R}^2\to\mathfrak{R}^2$ such that the flow curves in S are the curves

$$(t,x_2(t))=(t,x_2(0)+\xi(t)),\qquad 0\leqq t\leqq 1. \tag{3.22}$$

Let η be any C^1 function on $\mathfrak{R}^1$ that is periodic with period one and that is always positive. For $\mathbf{x}=(x_1,x_2)\in\mathfrak{R}^2$, set

$$b_1(\mathbf{x})=\eta(\xi(x_1)-x_2)\quad\text{and}\quad b_2(\mathbf{x})=\xi'(x_1)\eta(\xi(x_1)-x_2).$$

Then $\nabla\cdot\mathbf{b}(\mathbf{x})=0$ for $\mathbf{x}\in\mathfrak{R}^2$. Also $b_2(\mathbf{x})/b_1(\mathbf{x})=\xi'(x_1)$, and so the flow curves are given by (3.22). Note that every flow curve is periodic in the second component, i.e., $x_2(0)=x_2(1)$ for every flow curve. As a concrete example, take

$$b_1(\mathbf{x})=2+\sin(2\pi(\sin(2\pi x_1)-x_2)),\qquad b_2(\mathbf{x})=2\pi\cos(2\pi x_1)b_1(\mathbf{x}).$$

According to Theorem 3.10, each E_{ii} and K_{ii} is either $O(U_0^2)$ or $O(1)$. In fact, Proposition 3.9 implies that E_{11} and K_{11} are $O(U_0^2)$, and Corollary 3.8 implies that E_{22} and K_{22} are $O(1)$. It follows from the remarks after the proof of Lemma 3.3 that K_{12} is $o(U_0)$. With some effort, we can show that the inner products in (3.13) and (3.14) are zero, and so E_{12} and E_{21} are also $o(U_0)$.

Remark. Suppose the flow $Y(t, y)$ generated by $\mathbf{b} \cdot \nabla$ (i.e., $(d/dt)\dot{Y}(t, y) = \mathbf{b}(Y)$, $Y(0, y) = y$) is *ergodic* on T, with the normalized Lebesgue measure as the invariant measure. This is true if and only if the null space N is $\{0\}$. Since $b_i(Y(t, \cdot)) - \bar{b}_i$ is then ergodic, we may expect a smaller value of E_{ii} and, therefore, of the dispersion $K_{ii} = D_{ii} + 2E_{ii}$. Lemma 3.3 shows that this expectation is justified. The precise mathematical connection between the topological dynamics of $\mathbf{b}$ and the asymptotic behavior of the effective dispersion $\mathbf{K}(U_0)$, as $U_0 \to \infty$, appears complicated.

4. An asymptotic expansion of concentration. Assume that $D_{ij}(\cdot)$ and $b_i(\cdot)$ are continuously differentiable and periodic (with period one in each coordinate), $((D_{ij}(\cdot)))$ positive definite. Write $\dot{X}(t) = (X_1(t)(\text{mod } 1), \cdots, X_n(t)(\text{mod } 1))$. Then $\dot{X}(t)$ is a Markov process on the torus $[0, 1]^n$. Let $\dot{p}(t; \dot{x}, \dot{y})$ denote the transition probability density of $\dot{X}(t)$ and $\pi(\dot{y})$ the corresponding *invariant probability density*: $\int \pi(\dot{x})\dot{p}(t; \dot{x}, \dot{y})\, d\dot{x} = \pi(\dot{y})$. If the probability density of $\mathbf{X}(0)$ is π (the entire probability mass being on $[0, 1]^n$), then for any $t > 0$ the sequences $Y_j \equiv \mathbf{X}(jt) - \mathbf{X}((j-1)t)$ and $(Y_j, \dot{X}(jt))$ $(j = 1, 2, \cdots)$ are stationary and ϕ-mixing with an exponentially decaying ϕ-mixing rate, the latter being also Markovian (see Bhattacharya (1985)). Also, Y_j has a density and finite moments of all orders. Hence Theorem (2.8) of Götze and Hipp (1983) applies (see Example (1.13) in that article), and we have an asymptotic expansion for the distribution of $[\mathbf{X}(Nt) - \mathbf{X}(0) - NtU_0\bar{\mathbf{b}}]/N^{1/2} = [\sum_{j=1}^N (Y_j - EY_j)]/N^{1/2}$. More precisely we have, for every positive integer s,

$$\text{(4.1)} \quad \begin{aligned} &\text{Prob}\,((\mathbf{X}(Nt) - \mathbf{X}(0) - NtU_0\bar{\mathbf{b}})/N^{1/2} \in B) \\ &= \int_B \phi(t, \mathbf{x})\, d\mathbf{x} + \sum_{r=1}^{s} N^{-r/2} \int_B \psi_r(t, \mathbf{x})\, d\mathbf{x} + o(N^{-s/2}) \qquad (N \to \infty), \end{aligned}$$

uniformly over every class $\mathcal{B}$ of Borel sets B satisfying

$$\text{(4.2)} \quad \sup_{B \in \mathcal{B}} \int_{(\partial B)^\delta} \phi(t, \mathbf{x})\, d\mathbf{x} = O(\delta^a) \qquad (\delta \downarrow 0),$$

for some $a > 0$, $(\partial B)^\delta$ being the δ-neighborhood of the boundary ∂B of B. Here $\phi(t, \mathbf{x})$ is the Gaussian density with mean zero and dispersion matrix $t\mathbf{K}$, $\mathbf{K}$ being the large scale dispersion. The functions $\psi_r(t, \mathbf{x})$ are polynomial multiples of $\phi(t, \mathbf{x})$. For the classical case of independent summands the details of the construction of such polynomials may be found in Bhattacharya and Ranga Rao (1976, § 7). For the present case the formalism is entirely analogous once the cumulants of the normalized sum $\sum_1^N (Y_j - EY_j)/N^{1/2}$ are expanded in powers of $N^{-1/2}$ (see Götze and Hipp (1983)). Note that (4.2) holds, e.g., for the class of all Borel measurable convex sets (see Bhattacharya and Ranga Rao (1976, p. 24)).

In the case the initial concentration is proportional to π, (4.1) may be expressed as (see (1.6)),

$$\text{(4.3)} \quad \begin{aligned} &\int_B \varepsilon^{-n} C(\varepsilon^{-1}\mathbf{x} + \varepsilon^{-2}tU_0\bar{\mathbf{b}}, \varepsilon^{-2}t)\, d\mathbf{x} \\ &= C_0 \int_B \left[\phi(t, \mathbf{x}) + \sum_{r=1}^{s} \varepsilon^r \psi_r(t, \mathbf{x})\right] d\mathbf{x} + o(\varepsilon^s) \qquad (\varepsilon \downarrow 0), \end{aligned}$$

where C_0 is the total solute mass. On the other hand, if the initial concentration is arbitrary, say an integrable function or a point mass, the distribution of $\mathbf{X}(0)$ must be taken to be this concentration normalized. In this case Y_j is not stationary, but only asymptotically so, and the functions $\psi_r(t, \mathbf{x})$ must involve ε (or, $N^{-1/2}$) reflecting the nonstationarity of the moments, etc. Thus we have

$$\begin{aligned}\int_B \varepsilon^{-n} C(\varepsilon^{-1}\mathbf{x}+\varepsilon^{-2}tU_0\bar{\mathbf{b}}, \varepsilon^{-2}t)\, d\mathbf{x} \\ = C_0 \int_B \left[\phi(t,\mathbf{x}) + \sum_{r=1}^{s} \varepsilon^r \psi_r(t,\mathbf{x},\varepsilon)\right] d\mathbf{x} + o(\varepsilon^s) \qquad (\varepsilon \downarrow 0),\end{aligned} \tag{4.4}$$

uniformly over $B \in \mathscr{B}$ satisfying (4.2). It is very likely that (4.4) holds uniformly over the class of *all* Borel sets, i.e., the expansion holds in $L^1(\Re^n, d\mathbf{x})$; however, a proof of this does not seem to be available.

The expansion (4.4) provides a better approximation to concentration than the Gaussian approximation ϕ. This improvement is particularly significant for relatively small times, i.e., in the so-called preasymptotic zone. By computing the first three moments of observed concentration, we may approximately calculate the expansion (4.4) for $s = 1$. The fourth- and higher-order cumulants only contribute to terms $O(\varepsilon^2)$.

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