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On the method of moments for solute dispersion

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A cloud of solute injected into a pipe or channel is known to spread out by a dispersion process based on cross-sectional diffusion across a velocity shear. The original description of the process is due to Taylor (1953, 1954), and an important subsequent contribution was by Aris (1956), who framed and partially solved equations for the integral moments of the cloud of contaminant. The present work resolves some technical difficulties that occur when Aris' solution method (separation of variables) is pursued in depth. In particular, it is shown that Aris' technique has to be modified to give the moments at short and moderate times after the injection of solute into the flow. The paper is concerned with dispersion in those parallel flows for which an associated eigenvalue problem has a discrete spectrum of eigenvalues; fortunately, this case appears to be the rule rather than the exception. Expressions are obtained for the second and third moment about the mean, and the theory is applied to three cases of interest.

1. Introduction

The dispersion of soluble matter in fluid flow has been intensively researched since the classic papers on the subject by G. I. Taylor (1953, 1954). Taylor pointed out that solute is much more slowly dispersed by molecular or turbulent diffusion alone than by the process of molecular or turbulent diffusion across a velocity shear. This dispersion process has a wide application in chemical engineering and chromatography (see e.g. Marrero & Mason 1972; Howard 1976), environmental fluid mechanics (see e.g. Fischer *et al.* 1979) and other fields.

A complete mathematical description of the dispersion process is seldom possible, however, and approximation methods are invariably employed: for example, expansions that are valid for small (Barton 1978; Smith 1981) or large (Chatwin 1970) values of the time are available. Another fruitful approach is that developed by Aris (1956), namely the calculation of the first few integral moments of a cloud of solute as it disperses. The mean of the distribution and the second moment about the mean are particularly useful. It is now well known that a cloud of contaminant injected into a tube or channel develops a Gaussian profile in the downstream direction at asymptotically large times. Thus the dispersion process is asymptotically equivalent to a diffusion process, except that the apparent diffusion coefficient is much larger than ordinary molecular or turbulent diffusivities. The apparent diffusion coefficient may be readily calculated once the asymptotic form of the second moment about the mean is known: for example, Aris (1956) showed from the second moment that the dispersion coefficient is $\kappa + a^2U^2/48\kappa$ for a solute with molecular diffusivity κ injected into a tube of radius a containing fluid in laminar flow with mean speed U. The moments of a cloud of solute are easier to calculate than the concentration of the distribution itself, and Aris' techniques are therefore important and useful in many applications.

The aim of this paper is to resolve technical difficulties that occur when solving Aris' moment equations by separation of variables. Aris (1956, 1959) did not deal with these problems in his papers because he was concerned only with the *asymptotic* behaviour of the second moment about the mean; the problems occur when separation of variables is used to solve for the moments at *all times* after the injection of contaminant. The Aris moment equations consist of a parabolic partial differential equation giving the time-dependent cross-sectional structure and an ordinary differential equation for the evolution of cross-sectionally averaged moments. Some complete solutions of these equations are already available for particular cases (e.g. Chatwin 1970). The present work is more general, however, as it gives solutions at all times for the second and third moments of a cloud of solute injected into a parallel flow. The calculations presented for the third moment are restricted to the case when the injected solute is initially uniform across the cross-section. The solutions are also subject to the proviso that an eigenvalue problem that occurs in the separable solution possesses a discrete spectrum of eigenvalues. This proviso may be important for models of dispersion in parallel flows when the diffusion coefficient vanishes at the boundaries, although, even in these cases, the proviso does not automatically present a hindrance, as is shown in §6.

The contents of this paper are as follows. In §2, a summary is given of the Aris method of moments, and solutions for the second and third moments are obtained in §3. Then in §§4–6 the results are applied to three cases of some interest: dispersion of a solute with constant molecular diffusivity in plane Coutte flow and in Poiseuille flow, and dispersion in turbulent channel flow. The calculations for these three examples are simplified by assuming that the initial distribution of contaminant is uniform across the cross-section of the flow. The conclusions of the work are gathered together in §7. The most important of these is that Aris' results are correct at asymptotically large times, although his work could not describe the approach to the asymptotic state without the modifications contained herein.

2. The Aris moment equations

A summary of Aris' (1956) method of moments for a dispersing solute is given in this section. We consider a confined parallel flow in the direction of the axis Ox^* of Cartesian coordinates $Ox^*y^*z^*$. (An asterisk in this section denotes a dimensional constant or variable.) Define Ω^* to be the cross-section of the flow in the Oy^*z^* plane, and let $|\Omega^*|$ be its area and $\partial\Omega^*$ its boundary. Further, suppose the flow has velocity components $(u^*(y^*, z^*), 0, 0)$ and that a solute with diffusivity $\kappa^*(y^*, z^*)$ is injected into the flow. The diffusive flux of contaminant is given by $-\kappa^*\nabla^*C^*$, where $C^*(x^*, y^*, z^*, t^*)$ is the concentration.

It is convenient to work with dimensionless variables $\{x, y, z, t, u, K, C\}$ defined by

$$[x^*, y^*, z^*] = a^*[x, y, z], \tag{2.1 a}$$

$$t^* = a^{*2}t/D^*, \quad u^* = U^*u, \tag{2.1 b, c}$$

$$\kappa^* = D^*K, \quad C^* = Q^*C/a^{*3}\Omega, \tag{2.1 d, e}$$

where D^* and U^* are the mean values of κ^* and u^* over Ω^*, a^* is a characteristic scale of Ω^*, Q^* is the total amount of injected contaminant, and Ω is the dimensionless cross-sectional area of the flow. The factor Ω is included in the non-dimensionalization of C^* for convenience, and the definition ensures that $\iiint C\,dV = \Omega$. The dimensionless variables K and u have unit cross-sectional mean, that is

$$\bar{K} = \frac{1}{\Omega}\iint_{\Omega} K(y,z)\,dy\,dz = 1, \quad \bar{u} = \frac{1}{\Omega}\iint_{\Omega} u(y,z)\,dy\,dz = 1.$$

The concentration C then satisfies the equation

$$\frac{\partial C}{\partial t} + Pu\frac{\partial C}{\partial x} = K\frac{\partial^2 C}{\partial x^2} + \frac{\partial}{\partial y}\left(K\frac{\partial C}{\partial y}\right) + \frac{\partial}{\partial z}\left(K\frac{\partial C}{\partial z}\right) \tag{2.2}$$

under the conditions

$$K\nabla C\,.\,\hat{\mathbf{n}} = 0 \quad \text{at } \partial\Omega, \tag{2.3a}$$

$$C(x,\,y,\,z,\,0) = \mathscr{C}(x,\,y,\,z), \tag{2.3b}$$

$$\int_{-\infty}^{\infty} dx \iint_{\Omega} dy\,dz\,C = \Omega, \tag{2.3c}$$

$$C \text{ finite at all points,} \tag{2.3d}$$

$$x^n \frac{\partial^m C}{\partial x^m} \to 0 \quad \text{as } |x| \to \infty \quad (m,\,n = 0,1,2,\ldots). \tag{2.3e}$$

In (2.1), $p = U^*a^*/D^*$ is the Péclet number of the flow, and, in (2.3), $\hat{\mathbf{n}}$ is the unit normal to the boundary $\partial\Omega$ of the flow. The coordinates $Oxyz$ used in this work are stationary, whereas those used by Aris (1956) and Chatwin (1970) were moving with the discharge speed of the flow.

If the following moment definitions are now made,

$$C_n(y,\,z,\,t) = \int_{-\infty}^{\infty} x^n\,C(x,\,y,\,z,\,t)\,dx, \tag{2.4a}$$

$$M_n(t) = \bar{C}_n = \frac{1}{\Omega}\iint_{\Omega} C_n(y,\,z,\,t)\,dy\,dz, \tag{2.4b}$$

Aris has shown that C_n and M_n are the solutions of the problems

$$\frac{\partial C_n}{\partial t} - \frac{\partial}{\partial y}\left(K\frac{\partial C_n}{\partial y}\right) - \frac{\partial}{\partial z}\left(K\frac{\partial C_n}{\partial z}\right) = n(n-1)\,KC_{n-2} + nPuC_{n-1}, \tag{2.5a}$$

$$C_n(y,\,z,\,0) = \mathscr{C}_n(y,\,z), \tag{2.5b}$$

$$K\nabla C_n\,.\,\hat{\mathbf{n}} = 0 \quad \text{at} \quad \partial\Omega, \tag{2.5c}$$

$$C_n \text{ finite over the cross-section,} \tag{2.5d}$$

and

$$\frac{dM_n}{dt} = n(n-1)\,\overline{KC_{n-2}} + nP\,\overline{uC_{n-1}}, \tag{2.6a}$$

$$M_n(0) = \bar{\mathscr{C}}_n = \mathscr{M}_n. \tag{2.6b}$$

As previously, the overbar denotes the cross-sectional mean. The problems (2.5) and (2.6) are as defined by Aris, apart from minor changes of notation, the use of stationary coordinates and the explicit statement of the condition (2.5d). Aris has pointed out that the problems (2.5) and (2.6) can, in principle, be solved to as high a degree of accuracy as desired, and he was concerned with the asymptotic behaviour of $M_2(t)$. The parabolic partial differential equation (2.5a) can be solved by a variety of means: for example, by Laplace transform (Chatwin 1970) or by separation of variables (Aris 1956). The main thrust of the present paper is to obtain a solution for $M_0, \ldots, M_3$ using separation of variables, and, in doing so, to remedy errors implicit in Aris' formulation.

For later reference, the moments about the mean are also introduced at this point. They are defined by

$$\nu_n(t) = \frac{\iiint (x-\bar{x})^n C\,dv}{\iiint C\,dV},$$

where

$$\bar{x} = \frac{\iiint xC\,dV}{\iiint C\,dV} = \frac{M_1}{M_0},$$

and simple manipulations yield

$$\nu_2(t) = \frac{M_2}{M_0} - \bar{x}^2, \tag{2.7}$$

$$\begin{aligned} \nu_3(t) &= \frac{M_3}{M_0} - 3\bar{x}\frac{M_2}{M_0} + 2\bar{x}^2 \\ &= \frac{M_3}{M_0} - 3\bar{x}\nu_2 - \bar{x}^3. \end{aligned} \tag{2.8}$$

3. Solution of the moment equations by separation of variables

For $n = 0$, we look for a solution to (2.5) in the separable form

$$C_0 = A_{00} + \sum_i A_{0i} f_i\, e^{-\mu_i t}, \tag{3.1}$$

where the eigenvalue problem

$$\left\{\frac{\partial}{\partial y}\left(K\frac{\partial}{\partial y}\right) + \frac{\partial}{\partial z}\left(K\frac{\partial}{\partial z}\right) + \mu_i\right\} f_i = 0, \tag{3.2a}$$

$$K\nabla f_i \cdot \hat{\mathbf{n}} = 0 \quad \text{at } \partial\Omega, \quad f_i \text{ finite} \tag{3.2b}$$

is assumed to possess a discrete set of eigenvalues $\{\mu_i\}_{i=1}^{\infty}$ and corresponding eigenfunctions $\{f_i\}_{i=1}^{\infty}$ that are orthogonal and normalized so that

$$\bar{f}_i = 0, \quad \overline{f_i f_j} = \begin{Bmatrix} 1 & (i=j), \\ 0 & (i \neq j). \end{Bmatrix} \tag{3.3}$$

The eigenfunctions $\{f_i\}_{i=1}^{\infty}$ augmented by the constant 1 (with eigenvalue 0) form a complete set, and fitting the initial conditions $C_0(y, z, 0) = \mathscr{C}_0(y, z)$ fixes the constants A_{00} and A_{0i} to be

$$A_{00} = \overline{\mathscr{C}}_0, \quad A_{0i} = \overline{\mathscr{C}_0 f_i}. \tag{3.4}$$

Noting that $\mathscr{M}_0 = \overline{\mathscr{C}}_0 = 1$ by (2.3c), the solutions for M_0 and C_0 are

$$M_0(t) = 1, \tag{3.5a}$$

$$C_0(t) = 1 + \sum_i \overline{\mathscr{C}_0 f_i}\, f_i\, e^{-\mu_i t}. \tag{3.5b}$$

The solution for M_1 now follows readily by substituting for C_0 in (2.6). Without loss of generality, $\mathscr{M}_1$ is set equal to zero, and $M_1(t)$ is found to be

$$M_1(t) = Pt + P\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i}\,(1-e^{-\mu_i t})/\mu_i. \tag{3.6}$$

The term Pt reflects the well-known fact that the centre of mass ultimately moves at the discharge speed U^*, and the remaining terms give a small displacement relative to the original centre of mass.

The solution for C_1 is more complicated, however, and it is here that Aris' formulation would break down if expressions for the moments were required for other than asymptotically large times. The problem defining C_1 is

$$\frac{\partial C_1}{\partial t}-\frac{\partial}{\partial y}\left(K\frac{\partial C_1}{\partial y}\right)-\frac{\partial}{\partial z}\left(K\frac{\partial C_1}{\partial z}\right) = Pu+P\sum_i \overline{\mathscr{C}_0 f_i}\, uf_i\, e^{-\mu_i t}, \tag{3.7a}$$

$$C_1(y,z,0) = \mathscr{C}_1(y,z), \quad K\nabla C_1 \,.\, \hat{\mathbf{n}} = 0 \quad \text{at } \partial\Omega, \quad C_1 \text{ finite}, \tag{3.7b,c,d}$$

and, for reasons that are explained immediately below, the inhomogeneous terms in the defining equation are modified by adding in and subtracting out various terms as follows:

$$Pu+P\sum_i \overline{\mathscr{C}_0 f_i}\, uf_i\, e^{-\mu_i t} = P(u-\gamma_{10})+P\sum_i \overline{\mathscr{C}_0 f_i}\,(u-\gamma_{1i})f_i\, e^{-\mu_i t} + P\gamma_{10}+P\sum_i \overline{\mathscr{C}_0 f_i}\,\gamma_{1i} f_i\, e^{-\mu_i t}.$$

The solution for C_q is therefore

$$C_1 = A_{10}+\sum_i A_{1i} f_i\, e^{-\mu_i t}+P\phi_{10}+P\sum_i \overline{\mathscr{C}_0 f_i}\,\phi_{1i}\, e^{-\mu_i t}+Pt\gamma_{10}+Pt\sum_i \overline{\mathscr{C}_0 f_i}\,\gamma_{1i} f_i\, e^{-\mu_i t},$$

where the particular solutions ϕ_{10} and ϕ_{1i} are the solutions of the problems

$$\left\{\frac{\partial}{\partial y}\left(K\frac{\partial}{\partial y}\right)+\frac{\partial}{\partial z}\left(K\frac{\partial}{\partial z}\right)\right\}\phi_{10} = -(u-\gamma_{10}), \tag{3.8a}$$

$$K\nabla\phi_{10}\cdot\hat{\mathbf{n}} = 0 \quad \text{at } \partial\Omega, \quad \phi_{10} \text{ finite}, \tag{3.8b,c}$$

and

$$\left\{\frac{\partial}{\partial y}\left(K\frac{\partial}{\partial y}\right)+\frac{\partial}{\partial z}\left(K\frac{\partial}{\partial z}\right)+\mu_i\right\}\phi_{1i} = -(u-\gamma_{1i})f_i, \tag{3.9a}$$

$$K\nabla\phi_{1i}\,.\,\hat{\mathbf{n}} = 0 \quad \text{at } \partial\Omega, \quad \phi_{1i} \text{ finite}. \tag{3.9b,c}$$

Now it is well known from eigenvalue theory that inhomogeneous problems such as (3.8) and (3.9) possess a solution only if the inhomogeneous term is orthogonal to the eigenfunction of the homogeneous problem. This *solvability condition* (which was ignored by Aris for the equivalent of (3.9)) immediately determines the constants γ_{10} and γ_{1i} to be

$$\gamma_{10} = 1, \quad \gamma_{1i} = \overline{uf_i f_i}. \tag{3.10}$$

The introduction of the constants γ_{10} and γ_{1i} is essential to obtain the separable solution. Moreover, the solutions ϕ_{10} and ϕ_{1i} can be expressed as a linear combination of the complete set of eigenfunctions $\{1, \{f_i\}_{i=1}^{\infty}\}$. That is,

$$\phi_{10} = \beta^1_{00}+\sum_j \beta^1_{0j} f_j, \quad \phi_{1i} = \beta^1_{i0}+\sum_j \beta^1_{ij} f_j, \tag{3.11}$$

where the βs are determined by expanding the inhomogeneous terms in (3.8) and (3.9) in terms of the eigenfunctions. It is found that β^1_{00} and β^1_{ii} are arbitrary (and are hereinafter set equal to zero), and the other βs are given by

$$\beta^1_{0j} = \frac{\overline{uf_j}}{\mu_j}, \quad \beta^1_{i0} = -\frac{\overline{uf_i}}{\mu_i}, \tag{3.12a,b}$$

$$\beta^1_{ij} = -\frac{\overline{uf_i f_j}}{\mu_i-\mu_j} \quad (i \neq j). \tag{3.12c}$$

The solution of C_1 is completed by using the initial condition $C_1(y,z,0) = \mathscr{C}_1(y,z)$ to determine the arbitrary constants A_{10} and A_{1i} multiplying the complementary functions. This procedure is straightforward and, after some algebra, the final solution for C_1 is found to be

$$C_1 = Pt + P\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i}\,(1-e^{-\mu_i t})/\mu_i + \sum_i f_i\{\overline{\mathscr{C}_i f_i}\,e^{-\mu_i t} + P\overline{uf_i}\,(1-e^{-\mu_i t})/\mu_i\}$$
$$+P\sum_i\left\{\sum_{\substack{j\\ j\neq i}}\frac{\overline{uf_i f_j}}{\mu_j-\mu_i}(f_i\overline{\mathscr{C}_0 f_j}+f_j\overline{\mathscr{C}_0 f_i})\right\}e^{-\mu_i t} + Pt\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i f_i}f_i\,e^{-\mu_i t}. \quad (3.13)$$

The solution for M_2 may be obtained by substituting the expressions (3.5) and (3.13) for C_0 and C_1 into (2.6) and integrating. Omitting the lengthy details, M_2 is finally found to be

$$M_2(t) = \acute{\mathscr{M}}_2 + 2\{1+P^2\sum_i \overline{uf_i^2}/\mu_i\}t + 2P^2 t\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i}/\mu_i + P^2 t^2$$
$$-2P^2\sum_i \overline{uf_i^2}\,(1-e^{-\mu_i t})/\mu_i^2 + 2\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{Kf_i}\,(1-e^{-\mu_i t})/\mu_i$$
$$+2P\sum_i \overline{\mathscr{C}_1 f_i}\,\overline{uf_i}\,(1-e^{-\mu_i t})/\mu_i - 2P^2 t\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i}\,\overline{uf_i f_i}\,e^{-\mu_i t}/\mu_i$$
$$+2P^2\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i}\,\overline{(u-1)f_i f_i}\,(1-e^{-\mu_i t})/\mu_i^2$$
$$+2P^2\sum_i\left\{\sum_{\substack{j\\ j\neq i}}\frac{\overline{uf_i f_j}}{\mu_j-\mu_i}(\overline{\mathscr{C}_0 f_j}\,\overline{uf_i}+\overline{\mathscr{C}_0 f_i}\,\overline{uf_j})\right\}\frac{1-e^{-\mu_i t}}{\mu_i}. \quad (3.14)$$

This expression for M_2 enables $\nu_2(t)$ defined by (2.7) to be calculated. After simplification, $\nu_2(t)$ is found to be

$$\nu_2(t) = \mathscr{M}_2 + 2\{1+P^2\sum_i \overline{uf_i^2}/\mu_i\}t - 2P^2\sum_i \overline{uf_i^2}\,(1-e^{-\mu_i t})/\mu_i^2$$
$$+2\sum_i\left[\frac{1}{\mu_i}(\overline{\mathscr{C}_0 f_i}\,\overline{Kf_i} + P\overline{\mathscr{C}_1 f_i}\,\overline{uf_i}) + \frac{P^2}{\mu_i^2}\overline{\mathscr{C}_0 f_i}\,\overline{(u-1)f_i f_i}\right.$$
$$\left.+\frac{P^2}{\mu_i}\sum_j\frac{\overline{uf_i f_j}}{\mu_j-\mu_i}(\overline{\mathscr{C}_0 f_j}\,\overline{uf_i}+\overline{\mathscr{C}_0 f_i}\,\overline{uf_j})\right](1-e^{-\mu_i t})$$
$$-2P^2 t\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{(u-1)f_i f_i}\,\overline{uf_i}\,e^{-\mu_i t}/\mu_i - P^2(\sum_i \overline{\mathscr{C}_0 f_i}\,\overline{uf_i}\,(1-e^{-\mu_i t})/\mu_i)^2 \quad (3.15)$$

and, in the important case when the initial cloud of contaminant is uniform across the cross-section, this reduces to

$$\nu_2(t) = \mathscr{M}_2 + 2\{1+P^2\sum_i \overline{uf_i^2}/\mu_i\}t - 2P^2\sum_i \overline{uf_i^2}\,(1-e^{-\mu_i t})/\mu_i^2. \quad (3.16)$$

The moment $C_2(y, z, t)$ has to be found in order to calculate $M_3(t)$ and $\nu_3(t)$, and the analysis becomes very laborious at this point. For this exposition, it suffices to remark that inhomogeneous terms have to be added in and taken out of the defining equation for C_2 to satisfy solvability conditions. The particular solutions are then sought as a linear combination of the complete set $\{1, \{f_i\}_{i=1}^{\infty}\}$, and arbitrary constants are determined by fitting the initial condition $C_2(y, z, 0) = \mathscr{C}_2(y, z)$. Finally M_3 is found by substituting for C_1 and C_2 in (2.6) (with $n = 2$) and integrating. The resulting expression for M_3 is too long and complicated to reproduce here; rather the simplification that the initial cloud of contaminant is uniform across the channel is made for the presentation of results. In this case, the third moment about the mean is eventually found to be

$$\nu_3(t) = \mathscr{M}_3 + 6Pt \sum_i \frac{\overline{\mu f_i}}{\mu_i} (2\overline{Kf_i} + P^2 \sum_j \overline{uf_j}\,\overline{(u-1)f_i f_j}/\mu_j)$$

$$-12P \sum_i \frac{\overline{uf_i}}{\mu_i^2} \left\{ \overline{Kf_i} + P^2 \left[\frac{\overline{uf_i}\,\overline{(u-1)f_i f_i}}{\mu_i} - \sum_{\substack{j \\ j \neq i}} \frac{\overline{uf_j}\,\overline{uf_i f_j}}{\mu_i - \mu_j} \right] \right\} (1 - e^{-\mu_i t})$$

$$+ 6P^3 \sum_i \overline{(u-1)f_i f_i} \frac{\overline{uf_i^2}}{\mu_i^2} t e^{-\mu_i t}. \quad (3.17)$$

4. Dispersion in plane Couette flow

As a simple introductory example, we consider the dispersion of a solute with constant molecular diffusivity injected uniformly over the cross-section into plane Couette flow. In this case, K and u are given by $K = 1$, $u(y) = 2y$ $(0 \leqslant y \leqslant 1)$, and the eigenvalue problem (3.2) becomes

$$\left(\frac{d^2}{dy^2} + \mu_i\right) f_i = 0, \quad (4.1a)$$

$$\frac{df}{dy} = 0 \quad \text{at } y = 0, 1. \quad (4.1b)$$

The eigenvalues and corresponding normalized eigenvalues for this problem are therefore

$$\mu_i = (i\pi)^2, \quad f_i(y) = \sqrt{2} \cos i\pi y \quad (i = 1, 2, \ldots),$$

and it is an easy matter to derive the results

$$\overline{uf_i} = \frac{2\sqrt{2}\{(-1)^i - 1\}}{i^2\pi^2},$$

$$\overline{uf_i f_i} = 1,$$

$$\overline{uf_i f_j} = \frac{2}{\pi^2}\left\{\frac{(-1)^{i+j} - 1}{(i+j)^2} + \frac{(-1)^{i-j} - 1}{(i-j)^2}\right\},$$

which are required to calculate $\nu_2(t)$ and $\nu_3(t)$. If the various series in (3.16) and (3.17) are summed to 8 terms on a small calculator, $\nu_2(t)$ and $\nu_3(t)$ are found to be

$$\nu_2(t) = \mathscr{M}_2 - \frac{P^2}{148{\cdot}23529} + 2\left(1 + \frac{P^2}{30{\cdot}00003}\right)t + \frac{64P^2}{\pi^8} \sum_{i=1}^{\infty} \frac{1}{(2i-1)^8} e^{-(2i-1)^2\pi^2 t}, \quad (4.2a)$$

$$\nu_3(t) = \mathscr{M}_3. \quad (4.2b)$$

A simple check on the present theory is afforded by confirming that these expressions agree at large times with results predicted by Chatwin's asymptotic theory (1970, equations (3.8), (3.9)). The asymptotic theory predicts for ν_2 and ν_3

$$\nu_2(t) \sim \mathscr{M}_2 - \tfrac{17}{2520}P^2 + 2(1 + \tfrac{1}{30}P^2)t,$$

$$\nu_3(t) \sim \mathscr{M}_3,$$

and the agreements of the constants is excellent.

5. Dispersion in Poiseuille flow

This section considers the important case of a solute with constant molecular diffusivity injected uniformly across the cross-section into Poiseuille flow in a tube. The variables K and u are now given by

$$K = 1, \quad u(\rho) = 2(1-\rho^2) \quad (0 \leqslant \rho \leqslant 1), \tag{5.1}$$

where ρ is the usual cylindrical polar coordinate. (The assumption that $\mathscr{C}$ is uniform across the cross-section means that the analysis is independent of the other polar coordinate ϕ.) The eigenvalue problem (3.2) now becomes

$$\left\{\frac{d}{d\rho}\left(\rho\frac{d}{d\rho}\right)+\mu_i\rho\right\}f_i = 0, \tag{5.2a}$$

$$\frac{df_i}{d\rho} = 0 \quad \text{at} \quad \rho = 1, \quad f_i \text{ finite}, \tag{5.2b, c}$$

and the corresponding eigenvalues and normalized eigenfunctions for this problem are

$$\mu_i = \alpha_i, \quad f_i(\rho) = \frac{J_0(\alpha_i\rho)}{J_0(\alpha_i)}. \tag{5.3}$$

Here J_0 is the Bessel function of order 0, $\{\alpha_i\}_{i=1}^{\infty}$ are the roots of $J_0' = -J_1$, and ρ is the weight function in the orthogonality property

$$\frac{1}{\pi}\int_0^{2\pi} d\theta \int_0^1 \rho\frac{J_0(\alpha_i\rho)\,J_0(\alpha_j\rho)}{J_0(\alpha_i)\,J_0(\alpha_j)}d\rho = \begin{cases}1 & (i=j),\\ 0 & (i \neq j).\end{cases}$$

Some standard properties of Bessel functions can then be used to establish the results

$$\overline{uf_i} = -\frac{8}{\alpha_i^2},$$

$$\overline{uf_i f_i} = \tfrac{4}{3},$$

$$\overline{uf_i f_j} = -\frac{8(\alpha_i^2+\alpha_j^2)}{(\alpha_i^2-\alpha_j^2)^2} \quad (i \neq j),$$

and the task of summing various infinite series in (3.16) and (3.17) is simplified by using the following remarkable results due to Rayleigh (see Watson 1966, §15.51):

m	1	2	3	4	5	6
$\sum_{i=1}^{\infty} \alpha_i^{-2m}$	$\frac{1}{8}$	$\frac{1}{192}$	$\frac{1}{3072}$	$\frac{1}{460080}$	$\frac{13}{8847360}$	$\frac{11}{110100480}$

The expressions (3.16) and (3.17) then become

$$\nu_2(t) = \mathscr{M}_2 - \tfrac{1}{360}P^2 + 2(1+\tfrac{1}{48}P^2)t + 128P^2\sum_i \alpha_i^{-8}\,e^{-\alpha_i^2 t}, \tag{5.4}$$

$$\nu_3(t) = \mathscr{M}_3 + \tfrac{1}{480}P^3(t-\tfrac{17}{112}) + 128P^3\sum_i \alpha_i^{-12}(t\alpha_i^4+18\alpha_i^2-240)\,e^{-\alpha_i^2 t}. \tag{5.5}$$

These results are in agreement with those obtained by Chatwin (1970, eqns (4.9), (4.10)) using Laplace transforms, with the exception of the term

$$128P^3 t\sum_i \alpha_i^{-8}\,e^{-\alpha_i^2 t}$$

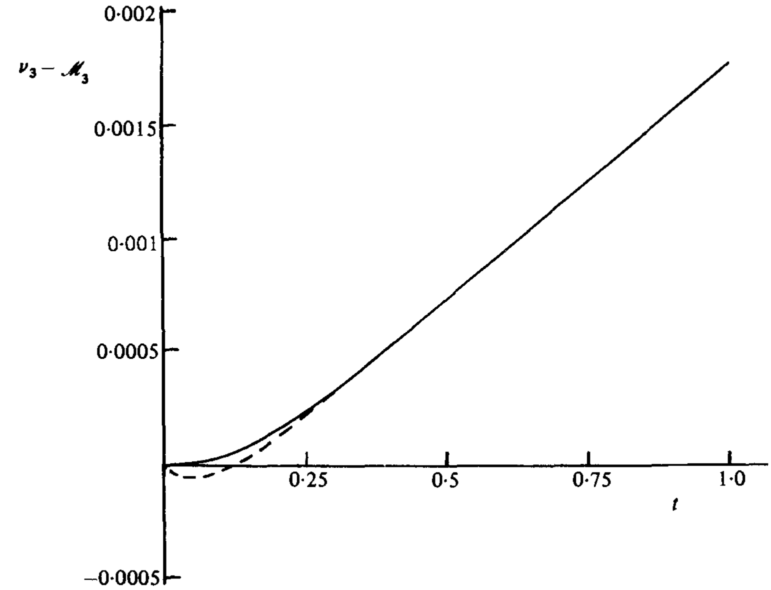

FIGURE 1. A comparison of $\nu_3(t)-\mathscr{M}_3$ for Poiseuille flow predicted by the present theory with Chatwin's (1970) result: ——, present theory; -----, Chatwin's result. The initial cloud of contaminant is uniform across the cross-section, and the variables are non-dimensionalized as in the text.

in $\nu_3(t)$. A close examination of appendix C of Chatwin's paper shows that this series results from neglected double zeros in a Laplace-transform inversion, and a comparison between the correct result (5.5) and Chatwin's result is shown in figure 1. To complete this section, it is noted that integral moments have been used by Andersson & Berglin (1981) to estimate diffusion coefficients from experiments in which solutes are dispersed in Poiseuille flow.

6. Dispersion in turbulent channel flow

Consider now, as a final application of the theory, the dispersion of a solute in turbulent channel flow. This subject has been studied by Elder (1959) and Chatwin (1970), who mentions that numerical solutions for the moments ν_2 and ν_3 have been given by W. W. Sayre (whose work is not readily accessible). Following Fischer *et al.* (1979, §4.2) the variables K and u are given by

$$K(y) = 6y(1-y), \quad u(y) = 1+\frac{u^*}{U^*\kappa}(1+\ln y) \quad (0 \leqslant y \leqslant 1), \tag{6.1}$$

in which u^* is the friction velocity, U^* is the discharge speed of the flow and κ is von Kármán's constant, which is approximately 0·42. As Chatwin (1970) points out, there are severe faults with the representations (6.1): there is no firm justification for the use of Reynolds' analogy, which gives $K(y)$ (but see Fischer *et al.* 1979, §5.1.1.1), the form for $u(y)$ neglects the important viscous sublayer (see Chatwin 1973), and turbulent flow in a channel is not two-dimensional. The theory also predicts results that differ by orders of magnitude from results in natural streams and watercourses (see Fischer *et al.* 1979, §5.2.1). In spite of these remarks, dispersion in turbulent flow in a channel is important, certainly for pedagogic reasons, and it is therefore included here.

$$f_1(y) = \surd 3(-1+2y)$$
$$f_2(y) = \surd 5(1-6y+6y^2)$$
$$f_3(y) = \surd 7(-1+12y-30y^2+20y^3)$$
$$f_4(y) = 3-60y+270y^2-420y^3+210y^4$$
$$f_5(y) = \surd 11(-1+30y-210y^2+560y^3-630y^4+252y^5)$$
$$f_6(y) = \surd 13(1-42y+420y^2-1680y^3+3150y^4-2772y^5+924y^6)$$
$$f_7(y) = \surd 15(-1+56y-756y^2+4200y^3-11550y^4+16632y^5-12012y^6+3432y^7)$$
$$f_8(y) = \surd 17(1-72y+1260y^2-9240y^3+34650y^4-72072y^5+84084y^6-51480y^7+12870y^8)$$
$$f_9(y) = \surd 19(-1+90y-1980y^2+18480y^3-90090y^4+252252y^5-420420y^6+411840y^7-218790y^8+48620y^9)$$

TABLE 1. The eigenfunctions $f_i(y)$ ($i = 1, \ldots, 9$) for dispersion in turbulent flow in the channel $0 \leqslant y \leqslant 1$

i	a_i	i	a_i
1	0·86603	11	0·03633
2	−0·37268	12	−0·03205
3	0·22048	13	0·02855
4	−0·15000	14	−0·02564
5	0·11055	15	0·02320
6	−0·08585	16	−0·02112
7	0·06916	17	0·01933
8	−0·05727	18	−0·01779
9	0·04843	19	0·01644
10	−0·04166		

TABLE 2. Values of the constants $a_i = \overline{f_i(y) \ln y}$

The eigenvalue problem (3.2) now becomes

$$\frac{d}{dy}\left(6y(1-y)\frac{df_i}{dy}\right)+\mu_i f_i = 0, \tag{6.2}$$

with

$$6y(1-y)\frac{df_i}{dy} = 0 \quad \text{at} \quad y = 0, 1, \tag{6.3}$$

which is equivalent to

$$f_i, f_i' \text{ finite} \quad \text{at} \quad y = 0, 1, \tag{6.4}$$

The problem specified by (6.2) and (6.4) is a singular Sturm–Liouville problem, and standard theory (see e.g. Boyce & Di Prima 1977) no longer guarantees that a discrete spectrum of eigenvalues exists. In the present case, however, there *is* a discrete spectrum, and the results developed in §3 still apply.

To show this, we make the substitution $\xi = 2y-1$, and (6.2) becomes Legendre's equation

$$\frac{d}{d\xi}\left((1-\xi^2)\frac{df_i}{d\xi}\right)+\tfrac{1}{6}\mu_i f_i = 0.$$

The only solutions of Legendre's equation that are finite at $\xi = \pm 1$ (and hence satisfy (6.4)) are the Legendre polynomials $P_n(\xi)$, and it merely remains to normalize these polynomials to satisfy the property $\overline{f_i f_i} = 1$. A computer program was written that generated the Legendre polynomials $P_n(\xi)$, sorted them into polynomials in y, and normalized them as in (3.3). The eigenfunctions $f_i(y)$ are displayed in table 1, and the various integrals $\overline{f_i(y) \ln y}$ and $\overline{f_i(y) f_j(y) \ln y}$ that are required for the application of the theory are given in tables 2 and 3. The eigenvalue corresponding to $f_i(y)$ is $\mu_i = 6i(i+1)$.

$i \backslash j$	1	2	3	4	5	6	7	8	9
1	−1·33333	0·96825	−0·45826	0·28868	−0·20516	0·15612	−0·12423	0·10202	−0·08579
2	c_{12}	−1·36667	0·98601	−0·47916	0·30901	−0·22395	0·17321	−0·13969	0·11603
3	c_{13}	c_{23}	−1·37619	0·99216	−0·48750	0·31798	−0·23289	0·18181	−0·14785
4	c_{14}	c_{24}	c_{34}	−1·38016	0·99499	−0·49167	0·32275	−0·23787	0·18681
5	c_{15}	c_{25}	c_{35}	c_{45}	−1·38218	0·99652	−0·49405	0·32559	−0·24095
6	c_{16}	c_{26}	c_{36}	c_{46}	c_{56}	−1·38334	0·99745	−0·49554	0·32742
7	c_{17}	c_{27}	c_{37}	c_{47}	c_{57}	c_{67}	−1·38408	0·99804	−0·49653
8	c_{18}	c_{28}	c_{38}	c_{48}	c_{58}	c_{68}	c_{78}	−1·38457	0·99846
9	c_{19}	c_{29}	c_{39}	c_{49}	c_{59}	c_{69}	c_{79}	c_{89}	−1·38491

TABLE 3. Values of the constants $c_{ij} = \overline{f_i(y) f_j(y) \ln y}$

To present the results, the constant $P(u^*/U^*\kappa)$ is first evaluated,

$$P(u^*/U^*\kappa) = \frac{U^*h^*}{D^*}\frac{u^*}{U^*\kappa} = \frac{U^*h^*}{\frac{1}{6}\kappa u^*h^*}\frac{u^*}{U^*\kappa} = \frac{6}{\kappa^2},$$

and the expressions (3.16) and (3.17) for $\nu_2(t)$ and $\nu_3(t)$ are then found to give

$$\nu_2(t) = \mathscr{M}_2 - \frac{0{\cdot}3835}{\kappa^4} + \left(2 + \frac{4{\cdot}849}{\kappa^4}\right)t + \frac{2}{\kappa^4}\sum_i \frac{a_i}{[i(i+1)]^2}e^{-\mu_i t}, \tag{6.5}$$

$$\nu_3(t) = \mathscr{M}_3 + t\left(\frac{1}{3\kappa^2} - \frac{4{\cdot}681}{\kappa^6}\right) + \frac{0{\cdot}6265}{\kappa^6} - \frac{1}{108\kappa^2}(1 - e^{-\mu_2 t})$$
$$+ \frac{2592}{\kappa^6}\sum_i \frac{a_i}{\mu_i^2}\left\{\frac{a_i(1+c_{ii})}{\mu_i} - \sum_{\substack{j \\ j\neq i}} \frac{a_j c_{ij}}{\mu_i - \mu_j}\right\}e^{-\mu_i t} + \frac{1296}{\kappa^6}\sum_i (1 + c_{ii})\frac{a_i^2}{\mu_i^2}t\, e^{-\mu_i t}. \tag{6.6}$$

Here μ_i, a_i and c_{ij} are given by

$$\mu_i = 6i(i+1), \quad a_i = \overline{f_i(y)\ln y}, \quad c_{ij} = \overline{f_i(y)f_j(y)\ln y},$$

and a_i and c_{ij} are tabulated. The asymptotic forms of these expressions agree well with the asymptotic forms quoted by Chatwin (1970, pp. 340–342), although there is a disagreement of about 5 % in the coefficient of t/κ^6 in $\nu_3(t)$. The numerical work for this section was carried out in double-precision arithmetic on a PDP 11/34 computer, with 20 terms retained in the various series; numerical accuracy to 3 figures should be assured. The results should be more accurate than those of Chatwin (1970). The expressions $\nu_2 - \mathscr{M}_2$ and $\nu_3 - \mathscr{M}_3$ are displayed in figure 2 with κ set equal to 0·42.

7. Discussion

The Aris method of moments is probably most useful in calculating the asymptotic form of the second moment about the mean of a dispersing cloud of solute. Such an expression provides the apparent diffusion coefficient for the dispersion process. In this respect, the errors implicit in Aris' (1956) paper are not important because they do not affect the leading asymptotic behaviour of $\nu_2(t)$. The solvability condition for (3.8) (which governs the asymptotic behaviour) is automatically satisfied provided the dispersion is described using coordinates moving with the mean speed of the flow. Thus Aris' results are adequate to give the asymptotic structure of $\nu_2(t)$ and $\nu_3(t)$, and his results are supported by the asymptotic results of Chatwin (1970).

The main result of the present paper is that expressions for $\nu_2(t)$ and $\nu_3(t)$ can be obtained using separation of variables, provided that due care is taken of solvability conditions in related eigenvalue problems. The new results are important if expressions for the moments are required at short or moderate times, or if information is required about the distribution of contaminant across the cross-section. It is found that the third moment is the highest obtainable with manageable labour and reasonable confidence in its accuracy. Fortunately, as Andersson & Berglin (1981) remark, this may often be sufficient even for use with accurate experiments.

The theory presented here should be capable of generalization to handle dispersion under different boundary conditions and in two-phase flows. The only requirements would be that a separable solution should exist and that the eigenvalue problem analogous to (3.2) should possess a discrete spectrum. The effect of changed boundary conditions might be particularly important: for example, there could be (possibly catalysed) reactions at the boundary (Barton 1982), or there could be a slow flux of

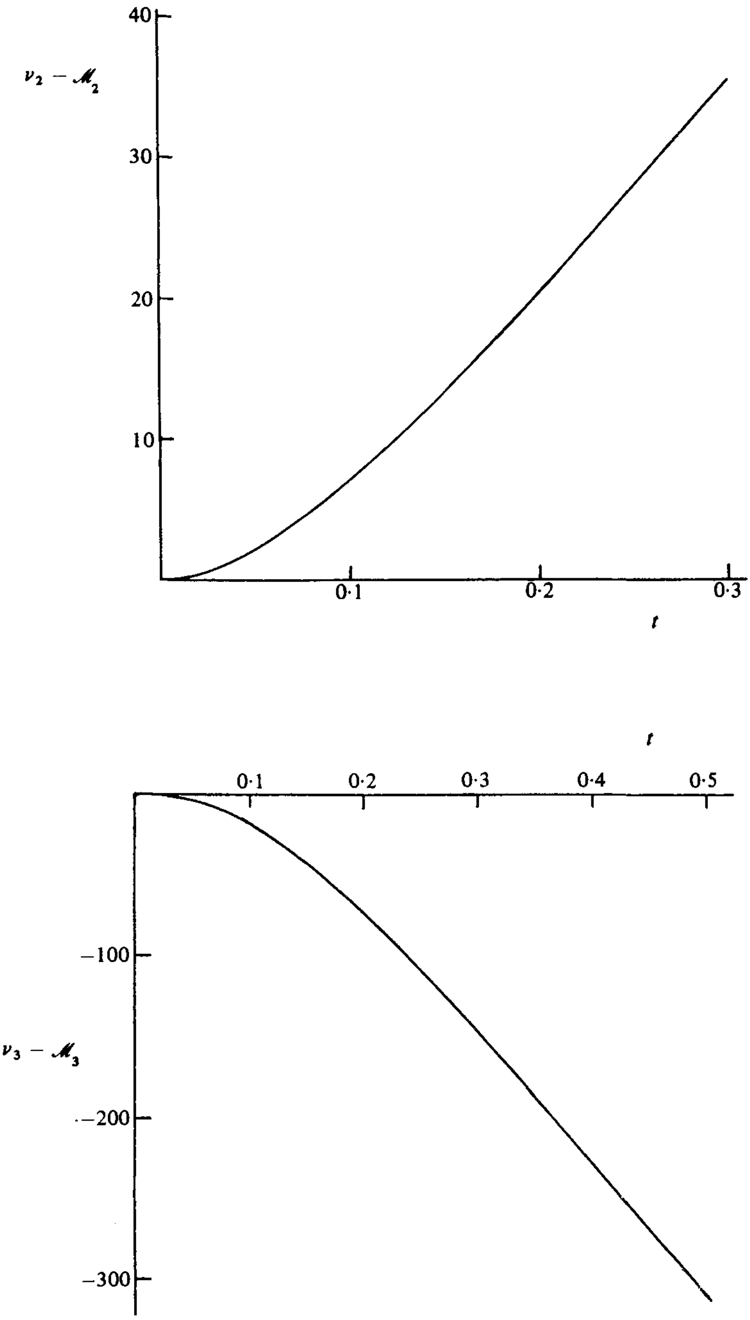

FIGURE 2. Graphs of $\nu_2(t) - \mathscr{M}_2$ and $\nu_3(t) - \mathscr{M}_3$ for dispersion in turbulent channel flow. The initial cloud of contaminant is uniform across the cross-section, and the variables are non-dimensionalized as in the text.

contaminant through the boundary. In these situations, the centre of mass of the contaminant cloud no longer moves at the discharge speed of the flow, and the solvability conditions are immediately required to determine the speed of the centre of mass and the apparent diffusion coefficient at large times. An analysis of this sort might find application to the situation described by Valentine & Wood (1977), where 'dead zones' at the edges of natural watercourses trap and then slowly release contaminant back into the flow.

Finally, when does (3.2) possess a discrete spectrum of eigenvalues? Standard eigenvalue theory (e.g. Boyce & Di Prima 1977) guarantees that a discrete spectrum exists if the problem is of normal Sturm–Liouville type – that is, if $K(y, z) > 0$, if the cross-section is of finite extent, and if the boundary condition is of the form

$$\alpha f_i + \beta \nabla f_i \cdot \hat{\mathbf{n}} = 0. \tag{7.1}$$

These conditions are automatically satisfied in the dispersion of a contaminant with constant molecular diffusivity in a laminar parallel flow through a finite cross-section. The conditions are not satisfied when modelling turbulent dispersion in a channel, as the eddy-diffusion coefficient vanishes at the boundaries. However, in the simple case analysed, a discrete spectrum did exist and the theory of §3 was applicable. It is conjectured that a discrete spectrum would still exist for this problem if the boundary conditions were changed from (6.3) and (6.4) to the form (7.1).

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