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The approach to normality of the concentration distribution of a solute in a solvent flowing along a straight pipe

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Taylor (1953, 1954*a*) showed that, when a cloud of solute is injected into a pipe through which a solvent is flowing, it spreads out, so that the distribution of concentration C is eventually a Gaussian function of distance along the pipe axis. This paper is concerned with the approach to this final form. An asymptotic series is derived for the distribution of concentration based on the assumption that the diffusion of solute obeys Fick's law. The first term is the Gaussian function, and succeeding terms describe the asymmetries and other deviations from normality observed in practice. The theory is applied to Poiseuille flow in a pipe of radius a and it is concluded that three terms of the series describe C satisfactorily if $Dt/a^2 > 0{\cdot}2$ (where D is the coefficient of molecular diffusion), and that the initial distribution of C has little effect on the approach to normality in most cases of practical importance. The predictions of the theory are compared with numerical work by Sayre (1968) for a simple model of turbulent open channel flow and show excellent agreement. The final section of the paper presents a second series derived from the first which involves only quantities which can be determined directly by integration from the observed values of C without knowledge of the velocity distribution or diffusivity. The latter series can be derived independently of the rest of the paper provided the cumulants of C tend to zero fast enough as $t \to \infty$, and it is suggested, therefore, that the latter series may be valid in flows for which Fick's law does not hold.

1. Introduction

This paper is concerned with the dispersion of a passive contaminant in a straight pipe of uniform cross-section, under the combined action of diffusion (molecular and/or turbulent) and advection with the fluid flowing along the pipe. It was shown by Taylor (1953, 1954 *a*) that for large times the mean concentration of contaminant over the cross-section satisfies a diffusion equation with respect to axes moving with the discharge velocity. Thus contaminant which is initially in the form of a cloud disperses so that its mean concentration is eventually a Gaussian function of distance along the pipe axis. Experiments by Taylor and others have substantially confirmed these conclusions, provided that sufficient time has elapsed since injection of the contaminant (e.g. Taylor 1953, figure 7).

The persistence of skewness and its causes

However, many experimental curves (e.g. Taylor 1953, figure 6) are not completely symmetric, because insufficient time has elapsed for asymmetries to be completely smoothed out. For cases in which the diffusion of the contaminant can be described by Fick's law, Aris (1956) has shown theoretically that the absolute skewness of the mean concentration does tend to zero, but only as $t^{-\frac{1}{2}}$, where t is the time since injection. Thus, any asymmetry that is present at any instant is likely to affect the observed concentration for a long time afterwards. There are at least two causes of such asymmetry being present.

First, the contaminant may be injected so that it is initially asymmetric. For example, the cloud of contaminant may have a long tail upstream or downstream. If the concentration of contaminant is $C(x,y,z,t)$ (where x measures distance along the axis, and y and z are co-ordinates in the cross-section), then the initial mean concentration will be asymmetric if ν_3 is initially different from zero, where, for $n > 1$,

$$\nu_n(t) = \iiint_{\substack{\text{whole}\\ \text{pipe}}} (x-x_g)^n\, C(x,y,z,t)\, dx\, dy\, dz \Big/ \iiint_{\substack{\text{whole}\\ \text{pipe}}} C(x,y,z,t)\, dx\, dy\, dz. \quad (1.1)$$

Thus, $\nu_n(t)$ is the nth integral moment of C. x_g is the x co-ordinate of the centre of mass of the cloud of contaminant.

Even if ν_3 is initially zero, it will, in general, rapidly become non-zero under the influence of advection with the fluid (which dominates over diffusion in the early stages of the dispersion; see Taylor 1953). Consider the simple model of turbulent flow in an open channel of depth h (figure 1 (a)), in which the mean flow velocity is logarithmic, and, with respect to axes moving with the discharge velocity, has the form

$$\frac{u_*}{\kappa}\left(1+\log\frac{y}{h}\right),$$

where u_* is the friction velocity and κ is von Kármán's constant. For a case when the cloud of contaminant is initially uniform over the depth and localized near $x = 0$, the concentration per unit width is approximately

$$C(x,y,0) = \frac{Q}{h}\,\delta(x),$$

where Q is the volume of contaminant per unit width. The cross-sectional mean of the concentration, $\bar{C}(x,0)$, is then identical with $C(x,y,0)$, and is sketched schematically in figure 1 (b). It is of course symmetric in the axial direction. Now, initially the cloud disperses under the influence of advection, so that, until diffusion is important,

$$C(x,y,t) = \frac{Q}{h}\,\delta\left[x-\frac{u_* t}{\kappa}\left(1+\log\frac{y}{h}\right)\right].$$

Evidently, $x_g = 0$, because of the choice of axes; and, by integrating, it is found that

$$\bar{C}(x,t) = \begin{cases} 0 & \text{if} \quad x > \dfrac{u_* t}{\kappa}, \\[2ex] \dfrac{Q\kappa}{h u_* t}\exp\left(\dfrac{\kappa x}{u_* t}-1\right) & \text{if} \quad x < \dfrac{u_* t}{\kappa}. \end{cases}$$

This is sketched schematically in figure 1 (*c*) and the profile is clearly asymmetric. It can easily be shown that

$$\nu_3 = -2(u_* t/\kappa)^3.$$

The most natural non-dimensional measure of asymmetry is the absolute skewness, λ_3, defined by

$$\lambda_3 = \nu_3/\nu_2^{\frac{3}{2}}, \qquad (1.2)$$

which, for the present example, is equal to -2.

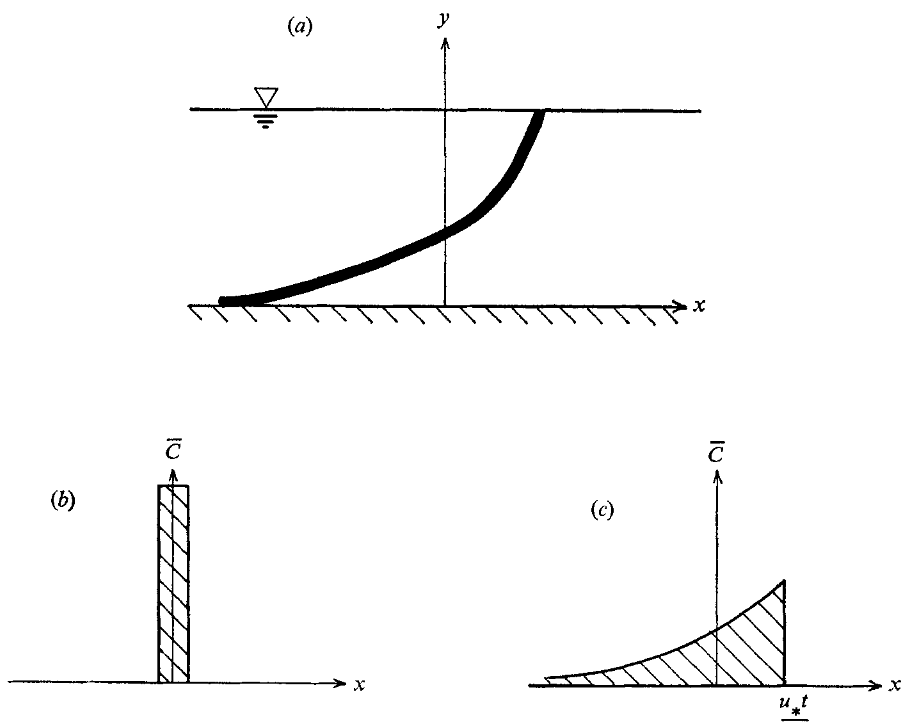

FIGURE 1. Initial dispersion of cloud of contaminant in turbulent open-channel flow, showing the asymmetry caused by advection.

Exceptionally, a particular velocity profile may be such that advection alone will not cause an initially symmetric distribution of concentration to become asymmetric. Poiseuille flow in a circular pipe of radius a is one example. The axial velocity referred to axes moving with the discharge velocity U has the form

$$U(1-2r^2/a^2),$$

where $r^2 = y^2+z^2$. An initial distribution of concentration of the form

$$C(x,y,z,0) = \bar{C}(x,0) = (Q/\pi a^2)\,\delta(x),$$

where Q the total volume of contaminant is changed by advection alone to the form shaded in figure 2 (*a*), viz.

$$C(x,y,z,t) = (Q/\pi a^2)\,\delta[x-Ut(1-2r^2/a^2)].$$

The corresponding value of $\bar{C}(x,t)$ is

$$\bar{C}(x,t) = \begin{cases} 0 & \text{if} \quad |x| > Ut, \\ \dfrac{Q}{2\pi a^2 Ut} & \text{if} \quad |x| < Ut. \end{cases}$$

This is symmetric (see figure 2(*b*)) because of the peculiar property of Poiseuille flow that the area of an annulus in which the velocity lies between two values u and $u+\delta u$ is independent of u and proportional to δu (Lighthill 1966). However, this is modified by diffusion into an asymmetric form. For near the cross-section AA (figure 2(*a*)) the presence of the wall means that diffusion transports fluid only inwards to a region where the velocity is greater than at the wall. Thus, the actual concentration near AA is lower than that predicted by a theory in which

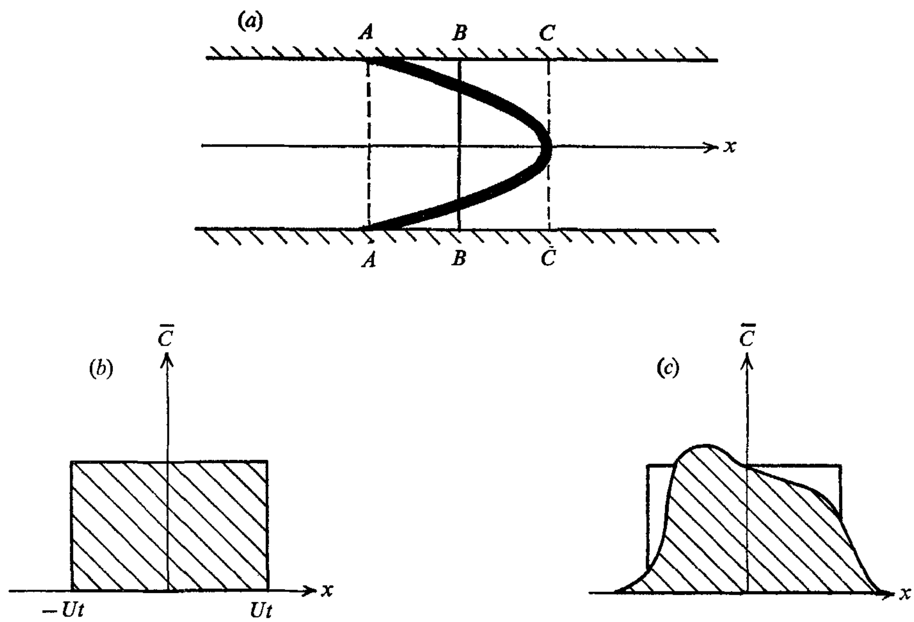

FIGURE 2. Initial dispersion of cloud of contaminant in Poiseuille flow, showing the asymmetry caused by diffusion and interaction with the pipe wall.

diffusion is ignored. The same thing is true near CC. However, the decrease in concentration here can be expected to be less than at AA, since, at cross-sections near to, but upstream of CC, some contaminant is diffused inwards to regions of higher velocity. Because the velocity gradient is zero on the pipe axis, the concentration becomes fairly uniform near the centre of the pipe in the neighbourhood of CC. This inward diffusion will therefore reduce, but not prevent, the lowering of the concentration at CC caused primarily by outward diffusion. Since the centre of gravity of the cloud of contaminant remains at BB (its position is not affected by diffusion), the actual form of $\bar{C}$ is likely to be asymmetric, of the type sketched schematically in figure 2(*c*).

Because asymmetries and other deviations from normality are known to be rather persistent, it is important that the following two problems be investigated:

(i) How does $\bar{C}$ approach a Gaussian form as $t \to \infty$?

(ii) How does the initial distribution of C influence this approach?

These questions are the motivation of the work in this paper.

Possible theoretical approaches

Attention will be given only to flows in which the diffusion of contaminant obeys Fick's law, i.e. to all flows in which the flux of contaminant can be written

$$-DK(y,z)\,\nabla C,$$

where D is a constant with the dimensions of diffusivity, and

$$\bar{K} = \frac{1}{S}\iint\limits_{\substack{\text{cross-}\\\text{section}}} K\,dy\,dz = 1, \tag{1.3}$$

where S is the total area of the cross-section. It will also be assumed that D and K are independent of concentration. Throughout this paper, an overbar on a quantity will denote its mean over the cross-section as in (1.3). Since the pipe is straight, the fluid velocity can be written

$$(UV(y,z),0,0),$$

where U is a characteristic velocity, and $\bar{V}$ is zero because the axes used are moving with the discharge velocity. The equation governing the concentration is thus

$$\frac{\partial C}{\partial t} + UV(y,z)\frac{\partial C}{\partial x} = DK\frac{\partial^2 C}{\partial x^2} + D\left[\frac{\partial}{\partial y}\left(K\frac{\partial C}{\partial y}\right) + \frac{\partial}{\partial z}\left(K\frac{\partial C}{\partial z}\right)\right]. \tag{1.4}$$

The boundary conditions on C are that

$$K\frac{\partial C}{\partial n} = 0 \quad \text{on pipe walls,} \tag{1.5}$$

since no contaminant flows through the walls, and that

$$C \to 0 \quad \text{as } |x| \to \infty, \tag{1.6}$$

if the contaminant is injected in the form of a cloud.† There is also an initial condition of the form,

$$C(x,y,z,0) = C^{(0)}(x,y,z). \tag{1.7}$$

An equation for $\bar{C}$ can be obtained by integrating (1.4) over the cross-section and using (1.5). This is

$$\frac{\partial \bar{C}}{\partial t} = -U\frac{\partial}{\partial x}\overline{VC} + D\frac{\partial^2}{\partial x^2}\overline{KC}. \tag{1.8}$$

† The theory of this paper can be adapted without difficulty to cover the case of transition from a region of uniform concentration C_1 to a region of uniform concentration C_2 for which the boundary condition (1.6) must be changed to: $C \to C_1$ as $x \to -\infty$, and $C \to C_2$ as $x \to \infty$. In this case, the infinite series (1.16) begins with a term independent of T.

There are several possible ways of investigating problems (i) and (ii). The first method considered was an extension of that used by Taylor (1953), who noticed that (1.4) has an exact steady solution in which C is linear in x. This solution can be written

$$C = \bar{C} + \frac{Ua^2}{D} g^{(1)}\left(\frac{y}{a}, \frac{z}{a}\right) \frac{d\bar{C}}{dx}, \tag{1.9}$$

where a is a characteristic length-scale of the cross-section, and

$$\frac{\partial}{\partial Y}\left(K\frac{\partial g^{(1)}}{\partial Y}\right) + \frac{\partial}{\partial Z}\left(K\frac{\partial g^{(1)}}{\partial Z}\right) = V \quad \text{with } Y = \frac{y}{a} \text{ and } Z = \frac{z}{a}. \tag{1.10}$$

The solution of (1.10) with the boundary condition (1.5) is not unique, but it becomes so if the further condition,

$$\overline{g^{(1)}} = 0, \tag{1.11}$$

is satisfied, and this condition is necessary if both sides of (1.9) are to have the same mean. Taylor now supposed that the above expression for C in terms of $\bar{C}$ remains approximately true even when $\partial^2\bar{C}/\partial x^2 \neq 0$, provided the cloud of contaminant is sufficiently elongated. On substituting (1.9) into (1.8), the following diffusion equation for $\bar{C}$ is obtained if a normally small term proportional to $\partial^3\bar{C}/\partial x^3$ is neglected

$$\frac{\partial\bar{C}}{\partial t} = \left\{D - \frac{U^2a^2}{D}\overline{Vg^{(1)}}\right\}\frac{\partial^2\bar{C}}{\partial x^2}. \tag{1.12}$$

The solution of (1.12) is the Gaussian curve,

$$\bar{C} \propto t^{-\frac{1}{2}} \exp\left\{-x^2 \Big/ 4t\left(D - \frac{U^2a^2}{D}\overline{Vg^{(1)}}\right)\right\}. \tag{1.13}$$

In order to obtain a better approximation it is natural to suppose, by analogy with (1.9), that $\bar{C}$ can be expressed in the form (Taylor 1954*b*; Gill 1967)

$$C = \bar{C} + \frac{Ua^2}{D} g^{(1)} \frac{\partial\bar{C}}{\partial x} + \left(\frac{Ua^2}{D}\right)^2 g^{(2)} \frac{\partial^2\bar{C}}{\partial x^2} + \ldots, \tag{1.14}$$

where $g^{(r)}$ is a function of Y and Z. When this expansion is substituted into (1.8), an equation for $\bar{C}$ is obtained, viz.

$$\frac{\partial\bar{C}}{\partial t} = \left\{D - \frac{U^2a^2}{D}\overline{Vg^{(1)}}\right\}\frac{\partial^2\bar{C}}{\partial x^2} + \left\{Ua^2\overline{Kg^{(1)}} - \frac{U^3a^4}{D^2}\overline{Vg^{(2)}}\right\}\frac{\partial^3\bar{C}}{\partial x^3} + \ldots. \tag{1.15}$$

It is possible to obtain the functions $g^{(r)}$ by substituting (1.14) into (1.4), and then using (1.15) to eliminate all time-derivatives. Equations for the $g^{(r)}$ are obtained by comparing coefficients of $\partial^r\bar{C}/\partial x^r$. The details of this procedure for Poiseuille flow are given in O'Hara (1969). The disadvantage of this method is that it does not give a direct expression for C or $\bar{C}$ until (1.15) has been solved to the required accuracy, and this will not be straightforward.

A second method is to determine, in the manner described by Aris (1956), the values of the integral moments of C, defined in (1.1). This technique has been widely used in numerical work (Sayre 1968), since a knowledge of the first few moments of C gives a great deal of information about C itself. Nevertheless, it has the same disadvantage as the first method. No direct expression is obtained for C, nor for $\bar{C}$.

The method finally decided upon arose from the belief that the difference between C (or $\bar{C}$) and the Gaussian curve (1.13) is $o(t^{-\frac{1}{2}})$ as $t \to \infty$ for a fixed value of $x/t^{\frac{1}{2}}$. This remark suggests that C can be expanded in an asymptotic series,

$$C \sim \frac{C^{(0)}(X, Y, Z)}{T} + \frac{C^{(1)}(X, Y, Z)}{T^2} + \frac{C^{(2)}(X, Y, Z)}{T^3} + \ldots, \tag{1.16}$$

where Y and Z are defined in (1.10), and

$$T = \left(\frac{MtD}{a^2}\right)^{\frac{1}{2}}, \quad X = x\left(\frac{D}{MU^2a^2t}\right)^{\frac{1}{2}}. \tag{1.17}$$

Here M is an arbitrary dimensionless constant whose value will be chosen later for algebraic convenience.

Plan of the paper

In § 2 it is shown that the above series is consistent with the equation for C, and expressions are obtained for the functions $C^{(p)}$. It is also shown that this expansion implies both (1.14) and (1.15). The work of § 3 is primarily concerned with the effect of the initial distribution of C on the terms in the series.

Some consequences of the theory for the widely studied case of Poiseuille flow in a circular pipe are discussed in § 4. In particular, graphs of $\bar{C}$, ν_2 and ν_3, obtained with the present theory, are compared with those obtained by previous writers, and an attempt is made to answer the important question of the range of times for which the asymptotic series is an adequate approximation.

Section 5 deals with consequences of the theory for the model of turbulent open-channel flow, discussed above, in which V is logarithmic and K parabolic. Comparisons are made with Sayre (1968).

One disadvantage of (1.16) for practical purposes is that the functions $C^{(p)}$ cannot be found unless V and K are known accurately. However, it is shown in § 6 that the asymptotic series for $\bar{C}$ can be transformed into another series involving only quantities, like the integral moments, that can be determined without knowledge of V or K by integration from the observed distribution of $\bar{C}$. The need for such a series has been emphasized by Sayre. It turns out that the new series is well-known in statistical theory, and that it is valid even if C does not satisfy a differential equation provided another requirement is met.

2. Derivation of the asymptotic series

When the expansion (1.16) is substituted into the equation for C, and the coefficients of $T^{-(p+1)}$ for each p are equated to zero, the following equations are obtained:

$$\frac{1}{T}\colon \quad \frac{\partial}{\partial Y}\left(K\frac{\partial C^{(0)}}{\partial Y}\right) + \frac{\partial}{\partial Z}\left(K\frac{\partial C^{(0)}}{\partial Z}\right) = 0, \tag{2.1}$$

$$\frac{1}{T^2}: \quad \frac{\partial}{\partial Y}\left(K\frac{\partial C^{(1)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial C^{(1)}}{\partial Z}\right) = V\frac{\partial C^{(0)}}{\partial X}, \tag{2.2}$$

$$\frac{1}{T^{p+1}}: \quad \frac{\partial}{\partial Y}\left(K\frac{\partial C^{(p)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial C^{(p)}}{\partial Z}\right)$$
$$= V\frac{\partial C^{(p-1)}}{\partial X}-\left(\frac{M}{2}\right)\left\{\left(\frac{D}{Ua}\right)^2\frac{2K}{M}\frac{\partial^2 C^{(p-2)}}{\partial X^2}+X\frac{\partial C^{(p-2)}}{\partial X}+(p-1)\,C^{(p-2)}\right\}. \tag{2.3}$$

Since the terms in the series are independent, each $C^{(p)}$ satisfies the same boundary conditions as C itself, viz.

$$\text{(i)}\quad K\frac{\partial C^{(p)}}{\partial n} = 0 \quad \text{on pipe walls,} \quad \text{(ii)}\quad C^{(p)}\to 0 \quad \text{as } |X|\to\infty. \tag{2.4}$$

The plan now is to determine the $C^{(p)}$ successively, beginning with $C^{(0)}$. A key property of the equation for $C^{(p)}$, used frequently in this procedure, is that it cannot be solved with the boundary condition (2.4) (i), unless the right-hand side of the equation for $C^{(p)}$, (2.3), has zero mean over the cross-section. This can be seen easily since, by Gauss's theorem and (2.4) (i),

$$\iint_S \left[\frac{\partial}{\partial Y}\left(K\frac{\partial C^{(p)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial C^{(p)}}{\partial Z}\right)\right]dY\,dZ = 0.$$

The same situation is well known for solutions of Poisson's equation with homogeneous Neumann boundary conditions. This property, applied to the equation for $C^{(p)}$, gives conditions on $C^{(p-1)}$ and $C^{(p-2)}$, which, when satisfied, reduce much apparent arbitrariness. In fact, it will be seen that each $C^{(p)}$ is determined to within one arbitrary constant, and in § 3 it will be shown how each of these arbitrary constants is determined by the initial distribution of C.

The equation for $C^{(0)}$ has the solution, satisfying (2.4) (i),

$$C^{(0)} = f^{(0)}(X), \tag{2.5}$$

where $f^{(0)}$ is so far arbitrary. The equation for $C^{(1)}$ is soluble, since $\overline{V} = 0$, and has as solution,

$$C^{(1)} = \frac{df^{(0)}}{dX}g^{(1)}(Y,Z)+f^{(1)}(X), \tag{2.6}$$

where $g^{(1)}$ is defined in (1.10) and (1.11), and $f^{(1)}(X)$ is so far arbitrary. Now, the equation for $C^{(2)}$ is, when expressions (2.5) and (2.6) are substituted,

$$\frac{\partial}{\partial Y}\left(K\frac{\partial C^{(2)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial C^{(2)}}{\partial Z}\right) = V\left\{g^{(1)}\frac{d^2f^{(0)}}{dX^2}+\frac{df^{(1)}}{dX}\right\}$$
$$-\frac{M}{2}\left\{\left(\frac{D}{Ua}\right)^2\frac{2K}{M}\frac{d^2f^{(0)}}{dX^2}+X\frac{df^{(0)}}{dX}+f^{(0)}\right\}.$$

As explained above, this is only soluble if the right-hand side has zero mean, so that

$$\frac{2}{M}\left\{\left(\frac{D}{Ua}\right)^2-\overline{Vg^{(1)}}\right\}\frac{d^2f^{(0)}}{dX^2}+X\frac{df^{(0)}}{dX}+f^{(0)} = 0. \tag{2.7}$$

If M, so far arbitrary, is chosen so that

$$M = 2\left\{\left(\frac{D}{Ua}\right)^2 - \overline{Vg^{(1)}}\right\}, \tag{2.8}$$

the solution of the equation for $f^{(0)}$, satisfying (2.4) (ii), is

$$f^{(0)} = \alpha^{(0,0)}\exp\left(-\tfrac{1}{2}X^2\right), \tag{2.9}$$

where $\alpha^{(0,0)}$ is a constant. Thus $C^{(0)}$ is determined, apart from the constant $\alpha^{(0,0)}$, and the equation for $C^{(2)}$ becomes

$$\frac{\partial}{\partial Y}\left(K\frac{\partial C^{(2)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial C^{(2)}}{\partial Z}\right) = V\frac{df^{(1)}}{dX}+\alpha^{(0,0)}\left\{Vg^{(1)}-\overline{Vg^{(1)}}-\left(\frac{D}{Ua}\right)^2(K-1)\right\}\frac{d^2}{dX^2}\exp\left(-\tfrac{1}{2}X^2\right).$$

It is useful in what follows to use the Hermite polynomials, $H_n(X)$, defined by

$$H_n(X)\exp\left(-\tfrac{1}{2}X^2\right) = (-1)^n\left(\frac{d}{dX}\right)^n\exp\left(-\tfrac{1}{2}X^2\right). \tag{2.10}$$

Thus,

$$\frac{\partial}{\partial Y}\left(K\frac{\partial C^{(2)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial C^{(2)}}{\partial Z}\right) = \alpha^{(0,0)}H_2\exp\left(-\tfrac{1}{2}X^2\right)\left\{Vg^{(1)}-\overline{Vg^{(1)}}-\left(\frac{D}{Ua}\right)^2(K-1)\right\}+V\frac{df^{(1)}}{dX},$$

with solution

$$C^{(2)} = \alpha^{(0,0)}H_2\exp\left(-\tfrac{1}{2}X^2\right)g^{(2)}(Y,Z)+\frac{df^{(1)}}{dX}g^{(1)}(Y,Z)+f^{(2)}(X), \tag{2.11}$$

where

$$\frac{\partial}{\partial Y}\left(K\frac{\partial g^{(2)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial g^{(2)}}{\partial Z}\right) = Vg^{(1)}-\overline{Vg^{(1)}}-\left(\frac{D}{Ua}\right)^2(K-1), \tag{2.12}$$

with (i) $K(\partial g^{(2)}/\partial n) = 0$ on pipe walls, (ii) $\overline{g^{(2)}} = 0$. (2.13)

$f^{(2)}(X)$ is at the moment arbitrary, but is determined by the integrability condition on the equation for $C^{(4)}$, in the same way as $f^{(0)}(X)$ is determined by the integrability condition on the equation for $C^{(2)}$.

The equation for $C^{(3)}$ can now be written down, and the condition for it to be soluble is a differential equation for $f^{(1)}(X)$, similar to that obtained above for $f^{(0)}(X)$. This equation has the solution,

$$f^{(1)} = \left\{\alpha^{(1,0)}H_1(X)+\frac{\alpha^{(0,0)}}{M}\left(\overline{Vg^{(2)}}-\left(\frac{D}{Ua}\right)^2\overline{Kg^{(1)}}\right)H_3(X)\right\}\exp\left(-\tfrac{1}{2}X^2\right), \tag{2.14}$$

where $\alpha^{(1,0)}$ is an arbitrary constant. Thus $C^{(1)}$ is determined apart from the constants $\alpha^{(0,0)}$ and $\alpha^{(1,0)}$, and the equation for $C^{(3)}$ can be solved in a similar fashion to that for $C^{(2)}$. The expression for $C^{(3)}$ involves a function $g^{(3)}(Y,Z)$, similar to $g^{(1)}$ and $g^{(2)}$, satisfying the equation,

$$\frac{\partial}{\partial Y}\left(K\frac{\partial g^{(3)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial g^{(3)}}{\partial Z}\right) = \{Vg^{(2)}-\overline{Vg^{(2)}}-\overline{Vg^{(1)}}g^{(1)}\}-\left(\frac{D}{Ua}\right)^2\{Kg^{(1)}-\overline{Kg^{(1)}}-g^{(1)}\}, \tag{2.15}$$

and boundary conditions like (2.13).

In order to avoid complicating the body of this paper with purely mathematical details, these are discussed in appendix A. It is sufficient to say here that, on continuing the procedure described above, the form of $C^{(p)}$ for any p can be seen and verified by induction. In particular, it is found that

$$
\begin{aligned}
C^{(0)} &= \{\alpha^{(0,0)}g^{(0)}\} H_0 \exp(-\tfrac{1}{2}X^2),\\
C^{(1)} &= \{\alpha^{(1,0)}g^{(0)} - \alpha^{(0,0)}g^{(1)}\} H_1 \exp(-\tfrac{1}{2}X^2) + \{\alpha^{(1,1)}g^{(0)}\} H_3 \exp(-\tfrac{1}{2}X^2),\\
C^{(2)} &= \{\alpha^{(2,0)}g^{(0)} - \alpha^{(1,0)}g^{(1)} + \alpha^{(0,0)}g^{(2)}\} H_2 \exp(-\tfrac{1}{2}X^2)\\
&\qquad + \{\alpha^{(2,1)}g^{(0)} - \alpha^{(1,1)}g^{(1)}\} H_4 \exp(-\tfrac{1}{2}X^2)\\
&\qquad + \{\alpha^{(2,2)}g^{(0)}\} H_6 \exp(-\tfrac{1}{2}X^2),\\
C^{(3)} &= \{\alpha^{(3,0)}g^{(0)} - \alpha^{(2,0)}g^{(1)} + \alpha^{(1,0)}g^{(2)} - \alpha^{(0,0)}g^{(3)}\} H_3 \exp(-\tfrac{1}{2}X^2)\\
&\qquad + \{\alpha^{(3,1)}g^{(0)} - \alpha^{(2,1)}g^{(1)} + \alpha^{(1,1)}g^{(2)}\} H_5 \exp(-\tfrac{1}{2}X^2)\\
&\qquad + \{\alpha^{(3,2)}g^{(0)} - \alpha^{(2,2)}g^{(1)}\} H_7 \exp(-\tfrac{1}{2}X^2)\\
&\qquad + \{\alpha^{(3,3)}g^{(0)}\} H_9 \exp(-\tfrac{1}{2}X^2).
\end{aligned} \tag{2.16}
$$

In these expressions $H_n(X)$ is the Hermite polynomial of degree n defined in (2.10) with $H_0 = 1$, and $g^{(1)}, g^{(2)}$ and $g^{(3)}$ are the functions of Y and Z defined above, with $g^{(0)} = 1$. Furthermore, the $\alpha^{(r,s)}$ are constants of which only the $\alpha^{(r,0)}$ are arbitrary. The $\alpha^{(r,s)}$ for $r \geqslant s \geqslant 1$ are linear functions of the $\alpha^{(r,0)}$ completely determined by the integrability conditions discussed above. In particular,

$$
\begin{aligned}
\alpha^{(1,1)} &= \alpha^{(0,0)}\left\{\frac{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}}{M}\right\},\\
\alpha^{(2,1)} &= -\alpha^{(0,0)}\left\{\frac{\overline{Vg^{(3)}} - (D/Ua)^2\,\overline{Kg^{(2)}}}{M}\right\} + \alpha^{(1,0)}\left\{\frac{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}}{M}\right\},\\
\alpha^{(2,2)} &= \frac{\alpha^{(0,0)}}{2}\left\{\frac{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}}{M}\right\}^2,\\
\alpha^{(3,1)} &= \alpha^{(0,0)}\left\{\frac{\overline{Vg^{(4)}} - (D/Ua)^2\,\overline{Kg^{(3)}}}{M}\right\} - \alpha^{(1,0)}\left\{\frac{\overline{Vg^{(3)}} - (D/Ua)^2\,\overline{Kg^{(2)}}}{M}\right\}\\
&\qquad + \alpha^{(2,0)}\left\{\frac{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}}{M}\right\},\\
\alpha^{(3,2)} &= -\alpha^{(0,0)}\frac{\{\overline{Vg^{(3)}} - (D/Ua)^2\,\overline{Kg^{(2)}}\}\{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}\}}{M^2}\\
&\qquad + \frac{\alpha^{(1,0)}}{2}\left\{\frac{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}}{M}\right\}^2,\\
\alpha^{(3,3)} &= \frac{\alpha^{(0,0)}}{6}\left\{\frac{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}}{M}\right\}^3.
\end{aligned} \tag{2.17}
$$

It only remains to determine the constants $\alpha^{(r,0)}$, and it is shown in §3 how this can be done.

Consistency with Taylor's work

Since
$$C \sim \frac{C^{(0)}}{T} + \frac{C^{(1)}}{T^2} + \frac{C^{(2)}}{T^3} + \ldots, \tag{2.18}$$
it follows that, as $T \to \infty$,
$$C = \bar{C} \sim \frac{C^{(0)}}{T} = \alpha^{(0,0)} \left(\frac{a^2}{MtD}\right)^{\frac{1}{2}} \exp\left\{-x^2\Big/4t\left(D - \frac{U^2a^2}{D}\,\overline{Vg^{(1)}}\right)\right\},$$
on returning to dimensional variables. This is exactly the expression (1.13) obtained by Taylor (1953) by the method discussed in §1.

It may be shown further that the asymptotic series is consistent with expressions (1.14) and (1.15). On substituting for $C^{(p)}$ in the asymptotic series and rearranging, it is found that
$$\begin{aligned} C \sim g^{(0)} &\left\{\frac{\alpha^{(0,0)}H_0 \exp(-\frac{1}{2}X^2)}{T} + \frac{(\alpha^{(1,0)}H_1 + \alpha^{(1,1)}H_3)\exp(-\frac{1}{2}X^2)}{T^2} + \ldots\right\} \\ &+ g^{(1)}\left\{\frac{\alpha^{(0,0)}H_1 \exp(-\frac{1}{2}X^2)}{T^2} + \frac{(\alpha^{(1,0)}H_2 + \alpha^{(1,1)}H_4)\exp(-\frac{1}{2}X^2)}{T^3} + \ldots\right\} \\ &+ \ldots. \end{aligned} \tag{2.19}$$
However, from (2.16) and (2.18),
$$\begin{aligned} \bar{C} \sim \frac{\alpha^{(0,0)}H_0 \exp(-\frac{1}{2}X^2)}{T} &+ \frac{(\alpha^{(1,0)}H_1 + \alpha^{(1,1)}H_3)\exp(-\frac{1}{2}X^2)}{T^2} \\ &+ \frac{(\alpha^{(2,0)}H_2 + \alpha^{(2,1)}H_4 + \alpha^{(2,2)}H_6)\exp(-\frac{1}{2}X^2)\ldots}{T^3}. \end{aligned} \tag{2.20}$$

Hence, combining (2.19) and (2.20), and returning to dimensional variables,
$$C \sim \bar{C} + \left(\frac{Ua^2}{D}\right) g^{(1)} \frac{\partial \bar{C}}{\partial x} + \left(\frac{Ua^2}{D}\right)^2 g^{(2)} \frac{\partial^2 \bar{C}}{\partial x^2} + \ldots,$$
which is simply (1.14). Equation (1.15) is a direct consequence of (1.14). For Poiseuille flow it has been verified that the coefficients in these series, obtained by the present method, agree with those found by O'Hara (1969). The details need not be given here.

3. Effect of initial distribution of C on its asymptotic form

The form of $\bar{C}$, shown in (2.20), gives a partial answer to question (i) of the introduction, how does $\bar{C}$ approach a Gaussian form as $t \to \infty$? The question cannot be regarded as completely answered until the $\alpha^{(r,0)}$ are determined. It is obvious that these constants must be determined by the initial distribution of C, since all other conditions that C must satisfy are met by the asymptotic series of §2, whatever the $\alpha^{(r,0)}$. Thus, as is to be expected, the complete answer to question (i) cannot be obtained until question (ii) is answered.

The method adopted here to determine the $\alpha^{(r,0)}$ is based on a consideration of the integral moments of C defined in (1.1). For large t these can be determined easily from the asymptotic series, using the orthonormal property of the Hermite polynomials,

$$\int_{-\infty}^{\infty} H_m H_n \exp(-\tfrac{1}{2}X^2)\,dX = (2\pi)^{\frac{1}{2}} m!\,\delta_{mn}, \tag{3.1}$$

and the following useful integral,

$$\int_{-\infty}^{\infty} X^m H_n \exp(-\tfrac{1}{2}X^2)\,dX = \begin{cases} 0 \quad \text{if} \quad m < n \quad \text{or} \quad (m-n) \quad \text{is odd}, \\ (2\pi)^{\frac{1}{2}} m! \dfrac{(m-n-1)(m-n-3)\ldots 1}{(m-n)!} \\ \qquad\qquad\qquad \text{if } (m-n) \text{ is even}. \end{cases} \tag{3.2}$$

Now, assuming the asymptotic series can be integrated term by term, and using the results above, it is easy to show that

$$\iiint C\,dx\,dy\,dz = S\int_{-\infty}^{\infty} \bar{C}\,dx = \alpha^{(0,0)}\left(\frac{Ua^2}{D}\right) S(2\pi)^{\frac{1}{2}}, \tag{3.3}$$

and

$$x_g = \frac{\iiint xC\,dx\,dy\,dz}{\iiint C\,dx\,dy\,dz} = \frac{\int_{-\infty}^{\infty} x\bar{C}\,dx}{\int_{-\infty}^{\infty} \bar{C}\,dx} = \left(\frac{\alpha^{(1,0)}}{\alpha^{(0,0)}}\right)\left(\frac{Ua^2}{D}\right). \tag{3.4}$$

Thus $\alpha^{(0,0)}$ is proportional to the total quantity of contaminant. Further, (3.4) shows that the centre of gravity of the cloud is asymptotically stationary, and it is possible, and convenient, to choose the origin of the co-ordinate system so that $x_g = 0$ asymptotically, Hence,

$$\alpha^{(1,0)} = 0, \tag{3.5}$$

and it follows, after some algebra, that

$$\nu_2(t) = \frac{\iiint (x-x_g)^2 C\,dx\,dy\,dz}{\iiint C\,dx\,dy\,dz} \sim \frac{MU^2a^2t}{D} + 2\left(\frac{\alpha^{(2,0)}}{\alpha^{(0,0)}}\right)\left(\frac{Ua^2}{D}\right)^2, \tag{3.6}$$

and

$$\nu_3(t) = \frac{\iiint (x-x_g)^3 C\,dx\,dy\,dz}{\iiint C\,dx\,dy\,dz} \sim 6\left(\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}\right)\left(\frac{MU^2a^2t}{D}\right)\left(\frac{Ua^2}{D}\right) + 6\left(\frac{\alpha^{(3,0)}}{\alpha^{(0,0)}}\right)\left(\frac{Ua^2}{D}\right)^3. \tag{3.7}$$

Here, $\alpha^{(1,1)}/\alpha^{(0,0)} = \{\overline{Vg^{(2)}} - (D/Ua)^2\,\overline{Kg^{(1)}}\}/M$, as shown above in (2.17). Taylor's (1953) theory gives $\nu_2 = MU^2a^2t/D$ and $\nu_3 = 0$. Thus, the present theory gives an additional contribution to the second moment which depends on the initial distribution of C, and a non-zero skewness with two contributions. The first of these is linear in time and independent of the initial distribution of C; the second is constant and does depend on the initial distribution of C.

The values of the integral moments can also be found from the equations developed by Aris (1956). It turns out that the values of the integral moments for large time can be obtained from these equations, in a form which shows the explicit dependence on the initial distribution of C. This is exactly what is required for the determination of the $\alpha^{(r,0)}$.

It is shown in appendix B how the asymptotic values of ν_2 and ν_3 can be obtained from the Aris equations. In particular, for an initial distribution of C that is uniform over the cross-section, it is found that, as $t \to \infty$,

$$\nu_2(t) \sim \frac{MU^2a^2t}{D} + \left\{\nu_2(0) - 2\left(\frac{Ua^2}{D}\right)^2 \overline{\{g^{(1)}\}^2}\right\}, \tag{3.8}$$

and

$$\nu_3(t) \sim 6\left(\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}\right)\left(\frac{MU^2a^2t}{D}\right)\left(\frac{Ua^2}{D}\right) + \left\{\nu_3(0) + 12\left(\frac{Ua^2}{D}\right)^3 \overline{g^{(1)}g^{(2)}}\right\}. \tag{3.9}$$

These expressions are consistent with (3.6) and (3.7) provided that

$$\frac{\alpha^{(2,0)}}{\alpha^{(0,0)}} = \frac{1}{2!}\left(\frac{D}{Ua}\right)^2 \frac{\nu_2(0)}{a^2} - \overline{\{g^{(1)}\}^2}, \tag{3.10}$$

and

$$\frac{\alpha^{(3,0)}}{\alpha^{(0,0)}} = \frac{1}{3!}\left(\frac{D}{Ua}\right)^3 \frac{\nu_3(0)}{a^3} + 2\overline{g^{(1)}g^{(2)}}. \tag{3.11}$$

These expressions show that the effect of the initial distribution of C on its asymptotic form is likely to be negligible for the common situation when $(Ua/D) \gg 1$, unless one or more moments are initially very large. This is an important conclusion, and explains why expressions for $\nu_2(t)$ and $\nu_3(t)$, when C is initially non-uniform over the cross-section, are not given in this paper.

The procedure outlined above can be used to determine the $\alpha^{(r,0)}$ for $r > 3$. Again, the details are not given here, because it will be argued later that the first three terms in the asymptotic series are usually an adequate approximation to $\bar{C}$.

4. Applications of the theory to Poiseuille flow in a circular pipe

The work of §2 and §3 has shown how the asymptotic form of C can be determined for any flow in which $V(Y,Z)$ and $K(Y,Z)$ are known. It is difficult to see immediately what the implications of the theory for any particular flow are. Therefore, in this section and the next, some of the applications of the theory to two typical flows are discussed. These applications are chosen so that the predictions of the theory can be compared with those made by others.

In this section the case of Poiseuille flow in a circular pipe is discussed. This example was that chosen by Taylor to illustrate his theory, and has since been used by Aris (1956) and Lighthill (1966), among others, to illustrate further aspects of the longitudinal dispersion process. Its great virtues are mathematical tractability and practical realizability.

For Poiseuille flow with discharge velocity U,

$$V(Y,Z) = (1-2R^2), \quad K(Y,Z) = 1,$$

where $R = (Y^2+Z^2)^{\frac{1}{2}}$. The functions $g^{(1)}$, $g^{(2)}$ and $g^{(3)}$ can be found from the defining equations (1.10), (2.12) and (2.15). They are

$$\left.\begin{aligned} g^{(1)} &= \frac{1}{24}(-2+6R^2-3R^4), \\ g^{(2)} &= \frac{1}{11,520}(31-180R^2+300R^4-200R^6+45R^8), \\ g^{(3)} &= \frac{1}{3,225,600}(109+490R^2-3185R^4+4900R^6-3500R^8 \\ &\qquad +1246R^{10}-175R^{12}). \end{aligned}\right\} \tag{4.1}$$

The values of these functions can be used to determine the constants $\alpha^{(r,s)}$, which occur in $C^{(0)}$, $C^{(1)}$ and $C^{(2)}$. As explained above, it is convenient to choose axes so that the centre of gravity of the cloud is at the origin as $t \to \infty$, for then $\alpha^{(1,0)} = 0$. Hence, using (2.17) and the expressions (4.1),

$$\frac{\alpha^{(2,1)}}{\alpha^{(0,0)}} = -\frac{1}{M}\frac{41}{2{,}580{,}480}, \quad \frac{\alpha^{(2,2)}}{\alpha^{(0,0)}} = \frac{1}{2M^2}\frac{1}{8{,}294{,}400}. \tag{4.2}$$

Furthermore, for a distribution of C that is initially independent of Y and Z, (3.10) and (3.11) give

$$\frac{\alpha^{(2,0)}}{\alpha^{(0,0)}} = \left(\frac{D}{Ua}\right)^2 \frac{\nu_2(0)}{2a^2} - \frac{1}{720}, \quad \frac{\alpha^{(3,0)}}{\alpha^{(0,0)}} = \left(\frac{D}{Ua}\right)^3 \frac{\nu_3(0)}{6a^3} - \frac{51}{967{,}680}. \tag{4.3}$$

The constant M is given by (2.8). For Poiseuille flow,

$$M = 2\left(\frac{D}{Ua}\right)^2 + \frac{1}{24}. \tag{4.4}$$

The asymptotic form of $\bar{C}$ when the initial distribution of concentration is uniform over the cross-section

The expressions above, when substituted into (2.20), give the asymptotic form of $\bar{C}$ for any value of Ua/D, provided the initial distribution of C is independent of Y and Z. However, for dispersion in liquids, Ua/D is normally very large (e.g. for $KMnO_4$ in water with $a = 0.1$ cm and $U = 1$ cm/sec, $Ua/D \sim 10^4$) so that the expressions above simplify.† In particular, $\alpha^{(2,0)}$ and $\alpha^{(3,0)}$ become independent of the initial form of C, and

$$M = \tfrac{1}{24}. \tag{4.5}$$

Hence, substituting in (2.20),

$$\bar{C} \sim \frac{\alpha^{(0,0)}\exp(-\frac{1}{2}X^2)}{T}\left[1 + \frac{1}{T}\frac{H_3(X)}{120} + \frac{1}{T^2}\left(-\frac{H_2(X)}{720} - \frac{41H_4(X)}{107{,}520} + \frac{H_6(X)}{28{,}800}\right) + \ldots\right], \tag{4.6}$$

where, from (1.17) and (4.5),

$$T = \left(\frac{Dt}{24a^2}\right)^{\frac{1}{2}}, \quad X = x\left(\frac{24D}{U^2a^2t}\right)^{\frac{1}{2}}. \tag{4.7}$$

The expression (4.6) can be written

$$\frac{\bar{C}}{\alpha^{(0,0)}(2\pi)^{\frac{1}{2}}} \sim \left[\frac{Z_0}{T} - \frac{Z_3}{120T^2} - \frac{1}{T^3}\left(\frac{Z_2}{720} + \frac{41Z_4}{107{,}520} - \frac{Z_6}{28{,}800}\right) + \ldots\right], \tag{4.8}$$

where
$$Z_n(X) = \frac{1}{(2\pi)^{\frac{1}{2}}}\left(\frac{d}{dX}\right)^n \exp(-\tfrac{1}{2}X^2).$$

† The assumption that $(D/Ua)^2$ is negligibly small means that the effect of longitudinal molecular diffusion on C is neglected compared with that due to the interaction between advection and lateral molecular diffusion. The flux of contaminant across a cross-section is of order $U(Ua^2/D)(\partial\bar{C}/\partial x)$ when longitudinal molecular diffusion is neglected (see (1.9)), whereas that due solely to the latter effect is of order $D(\partial\bar{C}/\partial x)$.

The Z_n are tabulated in Abramowitz & Stegun (1965); and table 1 gives the values of

$$Z_0, \ \frac{Z_3}{120} \quad \text{and} \quad \frac{Z_2}{720}+\frac{41Z_4}{107{,}520}-\frac{Z_6}{28{,}800}$$

for values of X between 0 and 5, and their values for negative X follow trivially, since Z_n is even if n is even, and odd if n is odd.

Figure 3 shows plots of $\surd(2/\pi)\,\bar{C}/\alpha^{(0,\,0)}$ against $Dx/2Ua^2$ given by those terms in (4.8) up to and including that of order T^{-3}, for four values of Dt/a^2 ranging from 0·125 to 1·000. For each value of Dt/a^2 the Gaussian curve given by Taylor's theory (and, therefore, by the first term in the series (4.8)) is shown for comparison. The units are chosen so that these graphs can be directly compared with those in Lighthill (1966).

X	0·00	0·20	0·50	1·00	1·50
1	$3{\cdot}99\times10^{-1}$	$3{\cdot}91\times10^{-1}$	$3{\cdot}52\times10^{-1}$	$2{\cdot}42\times10^{-1}$	$1{\cdot}30\times10^{-1}$
2	0	$1{\cdot}93\times10^{-3}$	$4{\cdot}03\times10^{-3}$	$4{\cdot}03\times10^{-3}$	$1{\cdot}21\times10^{-3}$
3	$1{\cdot}10\times10^{-4}$	$0{\cdot}71\times10^{-4}$	$-1{\cdot}00\times10^{-4}$	$-3{\cdot}18\times10^{-4}$	$-1{\cdot}41\times10^{-4}$

X	2·00	3·00	4·00	5·00
1	$5{\cdot}40\times10^{-2}$	$4{\cdot}43\times10^{-3}$	$1{\cdot}34\times10^{-4}$	$1{\cdot}49\times10^{-6}$
2	$-8{\cdot}93\times10^{-4}$	$-6{\cdot}65\times10^{-4}$	$-5{\cdot}79\times10^{-5}$	$-1{\cdot}36\times10^{-6}$
3	$1{\cdot}43\times10^{-4}$	$1{\cdot}15\times10^{-4}$	$6{\cdot}63\times10^{-6}$	$-0{\cdot}59\times10^{-7}$

TABLE 1. Values of the functions of X appearing in (4.8). Row 1 gives values of $Z_0(X)$. Row 2 gives values of $Z_3(X)/120$. Row 3 gives values of $Z_2(X)/720+41Z_4(X)/107{,}520-Z_6(X)/28{,}800$.

Leaving aside for the moment the question of the values of Dt/a^2 for which the series is an adequate approximation, it is worth noting several points:

(i) The asymmetry of $\bar{C}$ is evident for $Dt/a^2 = 0{\cdot}125$ but decreases as Dt/a^2 increases. However, it is still noticeable at $Dt/a^2 = 1{\cdot}000$. The asymmetry of $\bar{C}$ is due only to the term in Z_3 in the series (4.8), and it is this term which eventually gives the greatest correction to the Gaussian curve.

(ii) One effect of the term in (4.8) proportional to T^{-3} is to place the peak of $\bar{C}$ slightly behind the centre of gravity. This is consistent with the qualitative picture of figure 2(*c*).

(iii) For practical purposes there is no difference between the Gaussian curve and that given by (4.8) for values of Dt/a^2 greater than about 1.

(iv) The value of $\bar{C}$ predicted by the series falls off to zero more rapidly than the corresponding Gaussian curve as $|Dx/2Ua^2|$ increases. (Indeed, for $Dt/a^2 = 0{\cdot}125$, the series predicts that $\bar{C}$ is actually negative near $x = -Ut$!) This tendency is expected because, when longitudinal diffusion is neglected, the concentration cannot be non-zero upstream of a point moving with the maximum fluid velocity $+U$, nor downstream of a point moving with velocity $-U$. Thus $\bar{C} = 0$ for $|x| > Ut$, i.e. for $|Dx/2Ua^2| > Dt/2a^2$. The Gaussian curve does not satisfy this condition so that $\bar{C}$ must fall to zero more rapidly than the Gaussian curve.

Relation to work by Lighthill (1966)

The point discussed in (iv) above is among those considered by Lighthill (1966), who derived an expression for $\overline{C}$ valid for Dt/a^2 less than about 0·1 and for values of $Dx/2Ua^2 > 0$. The form of $\overline{C}$ given by Lighthill's theory falls off rapidly, from a uniform value, to reach zero at $x = Ut$. For values of $Dt/a^2 > 0{\cdot}1$, Lighthill

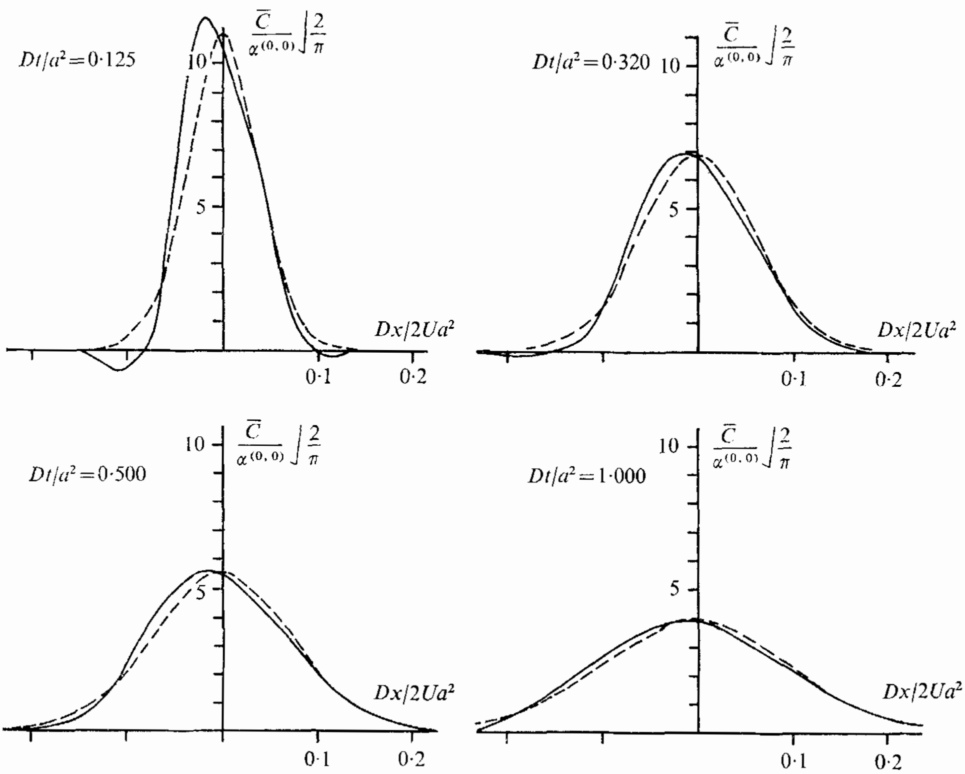

FIGURE 3. A comparison of $\overline{C}$ given by the present theory with that given by Taylor's theory for Poiseuille flow, and various times: ——, three terms of the present series; ------, the Gaussian curve of the same area.

suggested that the actual form of $\overline{C}$ lies between that predicted by his own and Taylor's theories, becoming indistinguishable from the latter near $Dt/a^2 = 0{\cdot}5$. Figure 4 shows the forms of $\overline{C}$ given by Lighthill's theory, Taylor's theory and the present theory for $Dt/a^2 = 0{\cdot}125$. Lighthill argues that his theory gives a peak value of $\overline{C}$ that is too low for $Dt/a^2 = 0{\cdot}125$, and that the Gaussian curve of Taylor's theory does not fall to zero rapidly enough.

For $Dt/a^2 = 0{\cdot}125$, the form of $\overline{C}$ predicted by the present theory has two main faults. First, $\overline{C}$ does not fall to zero rapidly enough, and secondly the curve is too 'spiky' near the centre of gravity (for, as shown in figure 2(*b*), the initial effect of Poiseuille flow is to give a $\overline{C}$ which is uniform for $|x| < Ut$).

Nevertheless, the curve given by the present theory is asymmetric and it must be so, as the argument of § 1 shows. Neither Taylor's nor Lighthill's theory predicts this, for Lighthill's theory is based on the fact that, for small values of Dt/a^2, the form of $\bar{C}$ near $|x| = Ut$ can be found by neglecting the presence of the pipe walls altogether. As has been seen in the introduction, it is just the interaction of the diffusing cloud with the pipe walls that causes the development of asymmetry in Poiseuille flow.

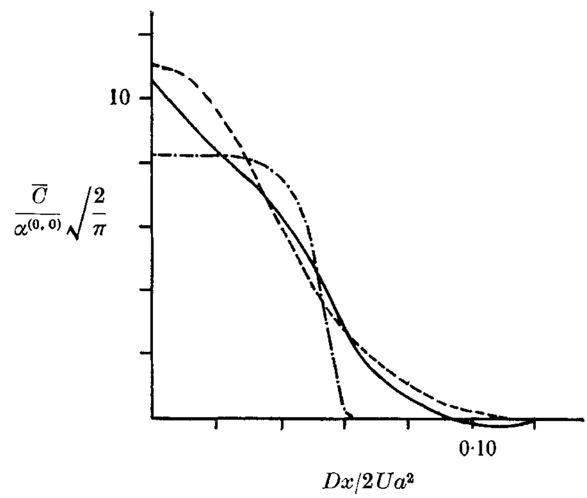

FIGURE 4. A comparison of the shapes of the $\bar{C}-x$ curves for Poiseuille flow with $Dt/a^2 = 0{\cdot}125$ given by various theories: ——, three terms of the present series; - - - - - -, the Gaussian curve of the same area; · —— · —— · ——, Lighthill's (1966) theory.

The second and third integral moments

The work above suggests that the present theory is not an adequate approximation for $Dt/a^2 = 0{\cdot}125$, but that it may be so for slightly higher values. In order to investigate this, the values of $\nu_2(t)$ and $\nu_3(t)$ given by the present theory are compared with the exact values given by solving the appropriate moment equations. For a case when

$$C(x,y,z,0) \propto \delta(x),$$

and when longitudinal molecular diffusion is neglected, it is shown in appendix C that

$$\left(\frac{D}{Ua}\right)^2 \frac{\nu_2(t)}{a^2} = \left[\frac{\frac{Dt}{a^2} - \frac{1}{15}}{24} + 128 \sum_{n=1}^{\infty} \frac{\exp(-\alpha_n^2 Dt/a^2)}{\alpha_n^8}\right], \tag{4.9}$$

$$\left(\frac{D}{Ua}\right)^3 \frac{\nu_3(t)}{a^3} = \left[\frac{\frac{Dt}{a^2} - \frac{17}{112}}{480} + 768 \sum_{n=1}^{\infty} \left(\frac{3\alpha_n^2 - 40}{\alpha_n^{12}}\right) \exp(-\alpha_n^2 Dt/a^2)\right], \tag{4.10}$$

where α_n is the nth non-zero root of $J_1(x) = 0$. The values of $\nu_2(t)$ and $\nu_3(t)$ given by the present theory are easily found from (3.8) and (3.9) to be the expressions (4.9) and (4.10) with the infinite series omitted. Figure 5 compares the exact values of ν_2 and ν_3 with those given by the series. Naturally, the theory agrees with the exact values as $Dt/a^2 \to \infty$, since such agreement has been demanded in finding the values of $\alpha^{(2,0)}$ and $\alpha^{(3,0)}$. But figure 5 shows that ν_2 and ν_3 are given with greater than 95 % accuracy by the asymptotic series for values of Dt/a^2 greater than about 0·25. Table 2 gives the exact and approximate values of the absolute skewness λ_3, defined in (1.2). Of course, the Gaussian curve has $\lambda_3 = 0$, whatever Dt/a^2.

Dt/a^2	0·02	0·05	0·10	0·15	0·20	0·50	1·00	5·00
Exact λ_3	−31·19	−3·97	−0·40	0·13	0·27	0·30	0·25	0·11
Approx. λ_3	—	—	−0·07	0·00	0·24	0·30	0·25	0·11

TABLE 2. Values of λ_3 in Poiseuille flow. A dash indicates that the present series gives a negative value of ν_2 so that λ_3 is not defined.

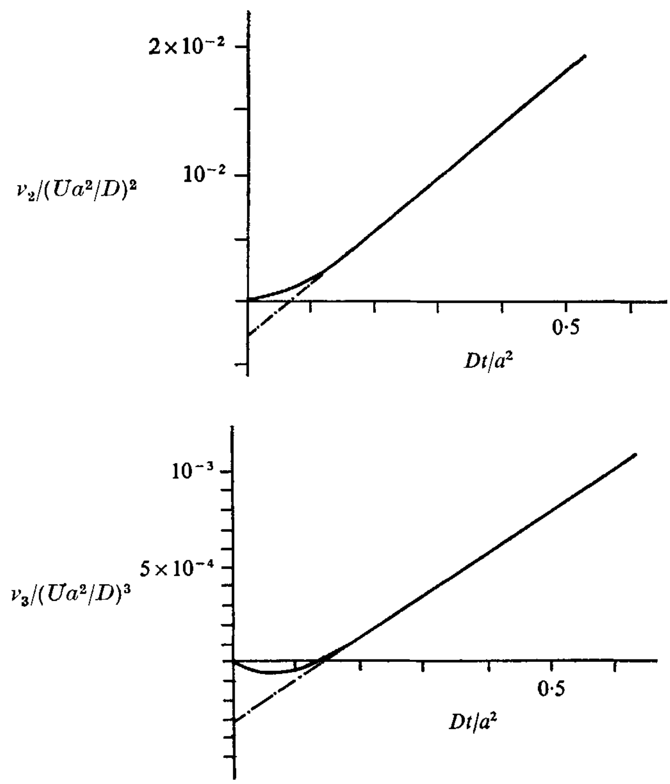

FIGURE 5. A comparison of the values of $\nu_2(t)$ and $\nu_3(t)$ for Poiseuille flow given by three terms of the present series with the exact values: ——, exact values; —·—, values given by three terms of the present series.

Concluding remarks

It is reasonable, therefore, to expect that in Poiseuille flow the first three terms of the asymptotic series (4.8) give an adequate approximation to $\bar{C}$ for values of Dt/a^2 greater than about 0·2. In particular, the series predicts the values of ν_2 and

ν_3 to within 95 % for $Dt/a^2 > 0{\cdot}25$. It is only in the event that information about higher moments than the third is required that more terms must be taken in the series.

5. Applications to a model of turbulent open channel flow

One of the commonest applications of the theory of longitudinal dispersion has been to turbulent flows in open channels, rivers and canals (Elder 1959; Fischer 1966; Sayre 1968). The simplest model of such a flow is discussed in § 1, and illustrated in figure 1. In this section, the velocity scale U and the length scale a of the general theory are chosen to be the friction velocity u_* and the depth of the channel h. Then, in the model,

$$V(Y,Z) = \frac{1}{\kappa}(1+\log Y), \quad \text{with} \quad Y = \frac{y}{h}, \tag{5.1}$$

where κ is von Kármán's constant equal to 0·42 approximately. It will also be assumed that the turbulent mixing can be described in terms of an eddy diffusivity, and that Reynolds's analogy holds. In this case,

$$D = \tfrac{1}{6}(\kappa u_* h) \quad \text{and} \quad K(Y,Z) = 6Y(1-Y). \tag{5.2}$$

There are many well-known faults with this model. The velocity near the wall in the viscous sublayer is not given by (5.1), there is no real justification for assuming an eddy diffusivity exists, let alone that Reynolds's analogy holds, and the motion in a channel is not independent of Z. However, the model is widely used by engineers, and numerical values of ν_2 and ν_3 are available for comparison with values predicted by the present theory (Sayre 1968).

Calculation of ν_2 and ν_3 from the theory

From (3.8), it follows that, as $t \to \infty$,

$$\nu_2(t) \sim \frac{6Mu_* ht}{\kappa} + \left\{\nu_2(0) - \frac{72h^2}{\kappa^2}\overline{\{g^{(1)}\}^2}\right\}, \tag{5.3}$$

where $g^{(1)}$ satisfies the appropriate form of (1.10), viz.

$$\frac{d}{dY}\left\{Y(1-Y)\frac{dg^{(1)}}{dY}\right\} = \frac{1}{6\kappa}\{1+\log Y\}, \tag{5.4}$$

and, from (2.8),

$$M = 2\left\{\frac{\kappa^2}{36} - \overline{Vg^{(1)}}\right\}. \tag{5.5}$$

The solution of (5.4) which satisfies $\overline{g^{(1)}} = 0$ is

$$g^{(1)} = \frac{1}{6\kappa}\left[1 + \int_0^Y \frac{\log u}{1-u}\,du\right]. \tag{5.6}$$

The integral occurring in (5.6) can be expressed in terms of the tabulated dilogarithm (Abramowitz & Stegun 1965). Table 3 gives the values of $6\kappa g^{(1)}$. From these values, the means $\overline{Vg^{(1)}}$ and $\overline{\{g^{(1)}\}^2}$ are found to be given approximately by

$$\overline{Vg^{(1)}} = -\frac{0\cdot 4041}{6\kappa^2} \quad \text{and} \quad \overline{\{g^{(1)}\}^2} = \frac{0\cdot 1885}{36\kappa^2}. \tag{5·7}$$

Y	$6\kappa g^{(1)}(Y)$	$36\kappa^2\{g^{(1)}(Y)\}^2$	$\frac{6\kappa Y\log^2 Y g^{(1)}(Y)}{(1-Y)}$
0·00	1·00000	1·00000	0·00000
0·05	0·75970	0·57714	0·35883
0·10	0·65478	0·42874	0·38573
0·15	0·53565	0·28692	0·34021
0·20	0·42986	0·18478	0·27837
0·25	0·33354	0·11125	0·21367
0·30	0·24444	0·05975	0·15186
0·35	0·16115	0·02597	0·09564
0·40	0·08265	0·00683	0·04626
0·45	0·00822	0·00007	0·00857
0·50	−0·06269	0·00393	−0·03012
0·55	−0·13054	0·01704	−0·05703
0·60	−0·19566	0·03828	−0·07659
0·65	−0·25832	0·06673	−0·08903
0·70	−0·31880	0·10163	−0·09460
0·75	−0·37728	0·14234	−0·09366
0·80	−0·43392	0·18829	−0·08644
0·85	−0·48890	0·23902	−0·07316
0·90	−0·54231	0·29410	−0·05418
0·95	−0·59428	0·35317	−0·02964
1·00	−0·64493	0·41593	0·00000

TABLE 3. Values of certain functions of Y used in the discussion of dispersion in turbulent open-channel flow. The values in the last column are used for the evaluation of

$$\overline{V\{g^{(1)}\}^2} = \frac{1}{\kappa}\int_0^1 (1+\log Y)\{g^{(1)}\}^2\, dY;$$

for, on integration by parts, it is seen that

$$\frac{1}{\kappa}\int_0^1 (1+\log Y)\{g^{(1)}\}^2\, dY = -\frac{1}{18\kappa^3}\int_0^1 \frac{Y\log^2 Y 6\kappa g^{(1)}}{(1-Y)}\, dY,$$

since

$$\frac{dg^{(1)}}{dY} = \frac{1}{6\kappa}\frac{Y}{1-Y}.$$

(In fact, the value of $\overline{Vg^{(1)}}$ can also be evaluated exactly in the manner described by Elder 1959.) Thus, from (5.3),

$$\frac{\nu_2(t)}{h^2} \sim 2\left(\frac{Dt}{h^2}\right) + \frac{4\cdot 849}{\kappa^4}\left(\frac{Dt}{h^2}\right) + \left\{\frac{\nu_2(0)}{h^2} - \frac{0\cdot 377}{\kappa^4}\right\}. \tag{5.8}$$

The first term in the expression is the effect of longitudinal diffusion, and the second is that given by Taylor's theory, resulting from the interaction between lateral diffusion and convection. The ratio of these two terms is

$$\frac{2\kappa^4}{4\cdot 849} = 0\cdot 0078 \approx 1\cdot 6\left(\frac{D}{u_* h}\right)^2.$$

As in Poiseuille flow, the ratio of the two terms is of the order of the inverse square of the Peclet number, and it is the interaction term which dominates. The other contributions to $\nu_2(t)$ are independent of time. One is the effect of the initial distribution of C (here, as in the general theory, assumed uniform over the cross-section), and the other is the result of the interaction between lateral diffusion and convection. Since $0{\cdot}377/\kappa^4 \approx 12$, it is the second term which is most important, unless the cloud of contaminant is initially very elongated.

For an initial distribution of contaminant of the form,

$$C(x,y,z,0) = \frac{Q}{h}\,\delta(x)\,\delta(z), \tag{5.9}$$

all moments initially vanish. Thus, as $t \to \infty$, and neglecting $2(Dt/h^2)$,

$$\frac{\nu_2(t)}{h^2} \sim \frac{4{\cdot}849}{\kappa^4}\left\{\frac{Dt}{h^2} - 0{\cdot}078\right\}. \tag{5.10}$$

Sayre (1968) evaluated $\nu_2(t)/h^2$ for this particular initial distribution on a computer. He found that for Dt/h^2 greater than about $0{\cdot}2$ the value of $\nu_2(t)/h^2$ was given by

$$\frac{\nu_2(t)}{h^2} \sim \frac{4{\cdot}85}{\kappa^4}\left\{\frac{Dt}{h^2} - 0{\cdot}079\right\}$$

(see figure (3.9) of Sayre's paper). The agreement between this and (5.10) is excellent.

One aspect of the expression (5.3) for ν_2 is worth mentioning in the context of turbulent diffusion theory. For an initial distribution of contaminant of the form (5.9), it follows that, as $t \to \infty$,

$$\nu_2(t) \sim \frac{6Mu_* h}{\kappa}\left\{t - \frac{12h}{Mu_*\kappa}\overline{\{g^{(1)}\}^2}\right\} = \frac{6Mu_* h}{\kappa}\{t - t_0\},$$

say, where $t_0 > 0$, since M and $\overline{\{g^{(1)}\}^2}$ are positive whatever V. However, the theory of turbulent diffusion predicts that, as $t \to \infty$,

$$\nu_2(t) \sim 2\int_0^\infty R(\xi)\,d\xi \left\{t - \frac{\int_0^\infty \xi R(\xi)\,d\xi}{\int_0^\infty R(\xi)\,d\xi}\right\},$$

where $R(\xi)$ is the statistical mean of the product of the velocities of a fluid particle at two times separated by an interval ξ (Batchelor 1966). Thus, the assumption of an eddy diffusivity implies a positive value of

$$\int_0^\infty \xi R(\xi)\,d\xi.$$

(The significance of

$$\int_0^\infty R(\xi)\,d\xi$$

in the present context is well known.)

A further check of the theory is provided by a calculation of $\nu_3(t)$ which, for large t, is given by (3.9) and (2.17). Neglecting the constant term,

$$\frac{\nu_3(t)}{h^3} \sim \frac{1296}{\kappa^3}\left\{\overline{Vg^{(2)}} - \frac{\kappa^2}{36}\overline{Kg^{(1)}}\right\}\left(\frac{Dt}{h^2}\right). \tag{5.11}$$

The value of $\{\overline{Vg^{(2)}} - (\kappa^2/36)\overline{Kg^{(1)}}\}$ can be found from the expression above for $g^{(1)}$, because, from (2.12), $g^{(2)}$ satisfies

$$\frac{d}{dY}\left\{6Y(1-Y)\frac{dg^{(2)}}{dY}\right\} = \{Vg^{(1)} - \overline{Vg^{(1)}}\} - \frac{\kappa^2}{36}\{K-1\}.$$

On multiplying this equation by $g^{(1)}$, integrating from 0 to 1, and integrating the left-hand side twice by parts, it follows that

$$\overline{Vg^{(2)}} = \overline{V\{g^{(1)}\}^2} - \frac{\kappa^2}{36}\{\overline{Kg^{(1)}}\}.$$

Thus,
$$\overline{Vg^{(2)}} - \frac{\kappa^2}{36}\overline{Kg^{(1)}} = \overline{V\{g^{(1)}\}^2} - \frac{\kappa^2}{18}\overline{Kg^{(1)}}.$$

The value of $\overline{Kg^{(1)}}$ is found exactly to be $-1/216\kappa$; and, from the values in table 3, it is found by Simpson's rule that

$$\overline{V\{g^{(1)}\}^2} = -0{\cdot}00346/\kappa^3.$$

Hence, from (5.11),

$$\frac{\nu_3(t)}{h^3} \sim \left(\frac{Dt}{h^2}\right)\left\{\frac{1}{3\kappa^2} - \frac{4{\cdot}486}{\kappa^6}\right\}. \tag{5.12}$$

Again the two terms in this expression represent respectively the effect of longitudinal diffusion and the interaction between lateral diffusion and convection, and it is the latter which is dominant. For large t, the value of the skewness λ_3, defined in (1.2), is

$$\lambda_3 = \frac{\nu_3}{\nu_2^{\frac{3}{2}}} \sim -\frac{0{\cdot}419}{(Dt/h^2)^{\frac{1}{2}}},$$

using (5.10) and (5.12). Sayre (1968) found that, for $Dt/h^2 \gg 0{\cdot}1$, λ_3 was approximately $-0{\cdot}42/(Dt/h^2)^{\frac{1}{2}}$. The agreement of the values of the constants is excellent.

Concluding remarks

The value of $\alpha^{(1,1)}$ has been found above so that, substituting in (2.20), viz.

$$\bar{C} \sim \frac{\alpha^{(0,0)}\exp(-\frac{1}{2}X^2)}{T}\left[1 + \frac{\alpha^{(1,1)}H_3(X)}{T} + O(T^{-2})\right];$$

and, writing as before
$$Z_n = \frac{1}{(2\pi)^{\frac{1}{2}}}\left(\frac{d}{dX}\right)^n \exp(-\tfrac{1}{2}X^2),$$

$$\frac{\bar{C}}{\alpha^{(0,0)}(2\pi)^{\frac{1}{2}}} \sim \left[\frac{Z_0}{T} - \frac{0{\cdot}062Z_3}{T^2} + O(T^{-3})\right], \tag{5.13}$$

where
$$T = 0{\cdot}87\left(\frac{Dt}{h^2}\right)^{\frac{1}{2}}, \quad X = 1{\cdot}14\left(\frac{Dx^2}{u_*^2h^2t}\right)^{\frac{1}{2}}. \tag{5.14}$$

Further terms in this series can be calculated by finding $g^{(2)}$ and $g^{(3)}$ in the manner described above. It seems likely, in view of the accuracy of ν_2 and ν_3, that three terms of the series will provide an adequate approximation to $\bar{C}$ for values of Dt/h^2 greater than about 0·2.

6. Expression of C by an Edgeworth series

The first three terms of the series (2.20) provide an adequate approximation to $\bar{C}$ for times which are not small. Unfortunately, the terms in this series cannot be evaluated if V and K are not known accurately, as occurs in many practical cases. It is the purpose of this section to show that the series (2.20) can be modified to one whose terms involve the integral moments, and these can be found by integration from the observed distribution of $\bar{C}$.

The first step in this modification is taken to simplify later algebra. The value of ν_2 for large t is given in (3.6), and it can be seen that a change in the origin of the time-scale can be made so that $\alpha^{(2,0)} = 0$, and

$$\nu_2(t) = \frac{MU^2a^2t}{D} = \left(\frac{Ua^2}{D}\right)^2 T^2. \tag{6.1}$$

With this change in the origin of the time-scale, the value of X, given by (1.17), can be written

$$X = x/\nu_2^{\frac{1}{2}}. \tag{6.2}$$

Thus, the change in the origin of the time-scale is equivalent to the statement that $\bar{C}$, regarded as a function of X, has zero mean and unit variance. It is this fact that causes simplification (as is well known in statistical theory).

When $\alpha^{(1,0)} = \alpha^{(2,0)} = 0$, the series (2.20) reduces to

$$\begin{aligned}\bar{C} \sim \frac{\alpha^{(0,0)}\exp\left(-\frac{1}{2}X^2\right)}{T}\Bigg[1 &+ \frac{1}{T}\left\{\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}H_3\right\} \\ &+ \frac{1}{T^2}\left\{\frac{\alpha^{(2,1)}}{\alpha^{(0,0)}}H_4 + \frac{\alpha^{(2,2)}}{\alpha^{(0,0)}}H_6\right\} \\ &+ \frac{1}{T^3}\left\{\frac{\alpha^{(3,0)}}{\alpha^{(0,0)}}H_3 + \frac{\alpha^{(3,1)}}{\alpha^{(0,0)}}H_5 + \frac{\alpha^{(3,2)}}{\alpha^{(0,0)}}H_7 + \frac{\alpha^{(3,3)}}{\alpha^{(0,0)}}H_9\right\} \\ &+ O(T^{-4})\Bigg].\end{aligned} \tag{6.3}$$

The plan now is to express the constants in this series in terms of the integral moments of $\bar{C}$. For example, the expression for ν_3, given in (3.7), shows that

$$\frac{1}{6}\left(\frac{\nu_3}{\nu_2^{\frac{3}{2}}}\right) = \left\{\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}\frac{1}{T} + \frac{\alpha^{(3,0)}}{\alpha^{(0,0)}}\frac{1}{T^3}\right\}. \tag{6.4}$$

Similarly, it is easy to show by integration that

$$\frac{1}{24}\left(\frac{\nu_4}{\nu_2^2} - 3\right) = \left\{\frac{\alpha^{(2,1)}}{\alpha^{(0,0)}}\frac{1}{T^2} + \frac{\alpha^{(4,0)}}{\alpha^{(0,0)}}\frac{1}{T^4}\right\}, \tag{6.5}$$

and

$$\frac{1}{120}\left(\frac{\nu_5}{\nu_2^{\frac{5}{2}}} - \frac{10\nu_3}{\nu_2^{\frac{3}{2}}}\right) = \left\{\frac{\alpha^{(3,1)}}{\alpha^{(0,0)}}\frac{1}{T^3} + \frac{\alpha^{(5,0)}}{\alpha^{(0,0)}}\frac{1}{T^5}\right\}. \tag{6.6}$$

The expressions on the right of (6.4,) (6.5) and (6.6) are respectively the coefficients of H_3, H_4 and H_5 in the series for $\bar{C}$. Furthermore, the combinations of the integral moments appearing on the left of these three equations are just those which define the absolute cumulants λ_n of $\bar{C}$. The definition of λ_3 has been given above in (1.2), and the definition of λ_n will be given later in this section. For the moment it is sufficient to note that

$$\lambda_3 = \frac{\nu_3}{\nu_2^{\frac{3}{2}}}, \quad \lambda_4 = \frac{\nu_4}{\nu_2^2} - 3, \quad \lambda_5 = \frac{\nu_5}{\nu_2^{\frac{5}{2}}} - \frac{10\nu_3}{\nu_2^{\frac{3}{2}}}, \tag{6.7}$$

and that the cumulants are particularly useful in measuring the departure of $\bar{C}$ from a Gaussian form, since all cumulants (except λ_2, which is 1) vanish for the latter.

The other constants appearing in the first few terms of (6.3) can also be expressed in terms of λ_3, λ_4 and λ_5. From (2.17) and (6.4),

$$\frac{\alpha^{(2,2)}}{\alpha^{(0,0)}}\frac{1}{T^2} = \frac{1}{2}\left\{\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}\frac{1}{T}\right\}^2 = \tfrac{1}{72}\lambda_3^2 + O(T^{-4}).$$

Similarly,

$$\frac{\alpha^{(3,2)}}{\alpha^{(0,0)}}\frac{1}{T^3} = \tfrac{1}{144}\lambda_3\lambda_4 + O(T^{-5}), \quad \frac{\alpha^{(3,3)}}{\alpha^{(0,0)}}\frac{1}{T^3} = \tfrac{1}{1296}\lambda_3^3 + O(T^{-5}).$$

Thus, substituting in (6.3),

$$\begin{aligned}\bar{C} \sim \frac{\alpha^{(0,0)}\exp\left(-\frac{1}{2}X^2\right)}{T}[1 &+ \{\tfrac{1}{6}\lambda_3 H_3\} + \{\tfrac{1}{24}\lambda_4 H_4 + \tfrac{1}{72}\lambda_3^2 H_6\} \\ &+ \{\tfrac{1}{120}\lambda_5 H_5 + \tfrac{1}{144}\lambda_3\lambda_4 H_7 + \tfrac{1}{1296}\lambda_3^3 H_9\} \\ &+ O(T^{-4})],\end{aligned} \tag{6.8}$$

where the terms in each curly bracket are of $O(T^{-1})$ times those in the preceding curly bracket.

The series (6.4) is well known in statistical theory as Edgeworth's form of the Gram–Charlier series of type A (Kendall & Stuart 1958, §6.18), and has been used in a discussion of sea waves by Longuet-Higgins (1963).

Alternative derivation of the Edgeworth series

The method of derivation of (6.4) relies heavily on the form of the series (2.20), which in turn relies on the assumption that the diffusion of contaminant can be described by a scalar diffusivity. Since this assumption is not known to be well founded for turbulent flow, an alternative derivation of (6.4), relying on different assumptions and very close to that used in statistical theory, may be of interest. The method is similar to that used by Aris (1958) for derivation of the Gram–Charlier series of type A, which, without Edgeworth's modification, has the disadvantage that the terms do not decrease regularly as T increases.

Define $\phi(k,t)$ as a Fourier transform of $\bar{C}(X,t)$, viz.

$$\phi(k,t) = \int_{-\infty}^{\infty} \bar{C}(X,t)\exp(ikX)\,dX \Big/ \int_{-\infty}^{\infty} \bar{C}(X,t)\,dX. \tag{6.9}$$

Thus, ϕ is the characteristic function of $\bar{C}$, and the cumulants λ_n of $\bar{C}$ are defined by the equation (Kendall & Stuart 1958, §3.12),

$$\log \phi(k,t) = \frac{(ik)^2}{2!} + \frac{(ik)^3}{3!}\lambda_3 + \ldots + \frac{(ik)^n}{n!}\lambda_n + \ldots. \tag{6.10}$$

The inverse of (6.9) is

$$\frac{\bar{C}(X,t)}{\int_{-\infty}^{\infty} \bar{C}\,dX} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \phi(k,t)\exp(-ikX)\,dk. \tag{6.11}$$

Hence, using (6.10),

$$\frac{\bar{C}(X,t)}{\int_{-\infty}^{\infty} \bar{C}\,dX} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp\left\{\frac{(ik)^3}{6}\lambda_3 + \frac{(ik)^4}{24}\lambda_4 + \frac{(ik)^5}{120}\lambda_5 + \ldots\right\}\exp(-ikX - \tfrac{1}{2}k^2)\,dk. \tag{6.12}$$

Now this expression can be simplified if the assumption is made, consistent with (6.4), (6.5) and (6.6), that, as $t \to \infty$,

$$\lambda_n = O(t^{1-\frac{1}{2}n}). \tag{6.13}$$

The exponential in the integrand of (6.12) can be expanded, and, grouping terms of the same order in t, it follows that

$$\frac{\bar{C}(X,t)}{\int_{-\infty}^{\infty} \bar{C}\,dX} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(-ikX - \tfrac{1}{2}k^2)\left[1 + \left\{\frac{(ik)^3}{6}\lambda_3\right\} + \left\{\frac{(ik)^4}{24}\lambda_4 + \frac{(ik)^6}{72}\lambda_3^2\right\} + \ldots\right]dk. \tag{6.14}$$

Thus, since

$$\int_{-\infty}^{\infty} \exp(-ikX - \tfrac{1}{2}k^2)\,(ik)^n\,dk = (2\pi)^{\frac{1}{2}} H_n(X)\exp(-\tfrac{1}{2}X^2),$$

and, from (3.3),

$$\int_{-\infty}^{\infty} \bar{C}\,dX = \frac{D}{Ua^2T}\int_{-\infty}^{\infty} \bar{C}\,dx = \alpha^{(0,0)}(2\pi)^{\frac{1}{2}}/T,$$

then (6.14) reduces to (6.8).

The attractive feature of this second derivation is that all the details involving V and the diffusion mechanism, needed to establish (2.20), are shown to be equivalent to the assumption (6.13). It can be shown that the values of the cumulants obtained from (2.20) are consistent for all n with (6.13), and it is natural to ask whether (6.13) can be justified for more general flows. So far, no such justification is known, but perhaps one may be forthcoming.

A further possibility is that the second derivation of the Edgeworth series may be adaptable to cases of diffusion in flows like turbulent jets in which the statistical properties of the velocity of a marked fluid particle are not constant.

Application of the Edgeworth series to practical flows

It is useful to outline the steps that are necessary to fit an observed distribution of $\bar{C}$ by the Edgeworth series.

(i) Determine the centre of gravity of $\bar{C}$ and choose axes so that this is zero.

(ii) Determine ν_2 by integration, and hence the values of X given by (6.2).

(iii) Determine ν_3, ν_4 and ν_5 by integration, and hence λ_3, λ_4 and λ_5 given in (6.7).

(iv) Determine Q, the total area under the $\bar{C}-x$ curve and hence $\alpha^{(0,0)}/T$, since

$$\alpha^{(0,0)}/T = Q/(2\pi\nu_2)^{\frac{1}{2}}.$$

All quantities appearing in (6.8) are now known.

This procedure has been applied to the observed distribution of $\bar{C}$ shown in figure 7 of Taylor (1954*a*), and the results are presented in table 4. The data in the first two columns of this table were read off the figure, it being assumed that $\bar{C}$ was zero at the extreme points recorded. The last three columns are the values

x	$\bar{C}$	First approx.	Second approx.	Third approx.
1·200	0·00	0·00	0·00	0·00
1·225	0·03	0·02	0·00	0·01
1·250	0·06	0·04	0·02	0·03
1·275	0·10	0·10	0·07	0·08
1·300	0·16	0·24	0·20	0·19
1·325	0·29	0·48	0·46	0·42
1·350	0·66	0·88	0·91	0·85
1·375	1·56	1·42	1·51	1·46
1·400	2·31	2·02	2·16	2·15
1·425	2·81	2·56	2·69	2·74
1·450	3·24	2·88	2·94	3·02
1·475	2·86	2·88	2·82	2·91
1·500	2·40	2·54	2·41	2·46
1·525	1·67	2·00	1·86	1·84
1·550	1·21	1·39	1·30	1·25
1·575	0·80	0·86	0·83	0·78
1·600	0·52	0·47	0·49	0·46
1·625	0·31	0·23	0·27	0·26
1·650	0·15	0·10	0·13	0·14
1·675	0·06	0·04	0·06	0·07
1·700	0·04	0·01	0·02	0·03
1·725	0·01	0·00	0·01	0·01
1·750	0·00	0·00	0·00	0·00

TABLE 4. Application of Edgeworth series to data from figure 7 of Taylor (1954*a*). Calculated values are $x_g = 1{\cdot}462$, $\nu_2^{\frac{1}{2}} = 0{\cdot}072$, $\lambda_3 = 0{\cdot}214$, $\lambda_4 = 0{\cdot}336$, $\alpha^{(0,0)}/T = 2{\cdot}924$.

calculated from (6.8), taking successively one, two and three terms in the series. It will be seen that the second and third approximations are clearly better than the first, although there are still points where agreement could be better. However, there are errors arising from experiment, errors made in reading values from the figure and errors arising because the curve shown in Taylor's paper is of $\bar{C}$ measured as a function of time as it passed the fixed place of observation, and not of $\bar{C}$ as a function of x for a given time. It is clearly necessary to test whether (6.8) provides an adequate approximation to $\bar{C}$ by considering more detailed sets of data; but these are not immediately available.

Appendix A. The values of $C^{(n)}(X, Y, Z)$

In § 2 the values of $C^{(n)}$ are given for $n = 0, 1, 2, 3$. The value of $C^{(n)}$ for arbitrary n is given by the natural extension of (2.16), and can be written

$$C^{(n)} = \sum_{s=0}^{n} g^{(n,s)}(Y, Z)\, H_{n+2s}(X) \exp\left(-\tfrac{1}{2}X^2\right), \tag{A 1}$$

where
$$g^{(n,s)} = \sum_{p=0}^{(n-s)} (-1)^p\, \alpha^{(n-p,s)} g^{(s)}(Y, Z). \tag{A 2}$$

The function $g^{(s)}(Y, Z)$ for arbitrary s satisfies an equation which is an extension of the equations defining $g^{(1)}$ and $g^{(2)}$, viz.

$$\begin{aligned}\frac{\partial}{\partial Y}\left(K\frac{\partial g^{(s)}}{\partial Y}\right) + \frac{\partial}{\partial Z}\left(K\frac{\partial g^{(s)}}{\partial Z}\right) &= \{Vg^{(s-1)} - \overline{Vg^{(s-1)}}g^{(0)} - \ldots - \overline{Vg^{(1)}}g^{(s-2)}\} \\ &\quad - (D/Ua)^2\{Kg^{(s-2)} - \overline{Kg^{(s-2)}}g^{(0)} - \ldots - \overline{Kg^{(0)}}g^{(s-2)}\} \\ &= \{Vg^{(s-1)} - (D/Ua)^2\, Kg^{(s-2)}\} \\ &\quad - \sum_{q=0}^{(s-1)} \left\{\overline{Vg^{(q)}} - \left(\frac{D}{Ua}\right)^2 \overline{Kg^{(q-1)}}\right\} g^{(s-q-1)},\end{aligned} \tag{A 3}$$

with the determining conditions,

$$\text{(i) } K(\partial g^{(s)}/\partial n) = 0 \quad \text{on pipe wall, and} \quad \text{(ii) } \overline{g^{(s)}} = 0 \quad \text{for} \quad s \geqslant 1. \tag{A 4}$$

The value of $g^{(0)}$ is of course 1.

As stated in the text, the constants $\alpha^{(r,s)}$ for $s \geqslant 1$ are linear functions of the $\alpha^{(r,0)}$ and are determined by the integrability conditions on the equations for the $C^{(r)}$. The values of $\alpha^{(r,s)}$ for $1 \leqslant r, s \leqslant 3$ are given in (2.17). For all integers r and s, the value of $\alpha^{(r,s)}$ can be written down as follows. Define the constants $\beta^{(q)}$ by

$$\beta^{(q)} = \overline{Vg^{(q)}} - \left(\frac{D}{Ua}\right)^2 \overline{Kg^{(q-1)}}. \tag{A 5}$$

Then, for $r \geqslant s \geqslant 1$,

$$\alpha^{(r,s)} = \frac{1}{s!\,M^s} \sum_{m=0}^{(r-s)} (-1)^m\, \alpha^{(r-s-m,0)} \{\Sigma \beta^{(q_1)} \beta^{(q_2)} \ldots \beta^{(q_s)}\}, \tag{A 6}$$

where the summation in the curly brackets is over *all* q_i such that, given m,

$$\text{(i)} \quad \sum_{i=1}^{s} q_i = 2s + m, \quad \text{and} \quad \text{(ii) } 2 \leqslant q_i \leqslant r - s + 2. \tag{A 7}$$

Thus, for example,

$$\alpha^{(4,2)} = \frac{1}{2M^2}\{\alpha^{(2,0)}[\beta^{(2)}]^2 - 2\alpha^{(1,0)}[\beta^{(2)}\beta^{(3)}] + \alpha^{(0,0)}[[\beta^{(3)}]^2 + 2[\beta^{(2)}\beta^{(4)}]]\}. \tag{A 8}$$

All the results given here can be verified by induction.

Appendix B. The values of ν_2 and ν_3 as $t \to \infty$

This appendix shows how the results (3.8) and (3.9) can be obtained. Let the Laplace transform of $C(x,y,z,t)$ be $\hat{C}(x,y,z,p)$. Then $\hat{C}$ satisfies the following equation obtained by transforming (1.4):

$$\frac{\partial}{\partial Y}\left(K\frac{\partial \hat{C}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial \hat{C}}{\partial Z}\right)+Ka^2\frac{\partial^2 \hat{C}}{\partial x^2} = \frac{Ua^2}{D}V\frac{\partial \hat{C}}{\partial x}+\frac{a^2}{D}(p\hat{C}-\mathscr{C}^{(0)}), \quad \text{(B 1)}$$

where Y and Z are defined in (1.10), and $\mathscr{C}^{(0)}$ is the initial distribution of concentration. Now, let $\hat{\hat{C}}$ be the Fourier transform of $\hat{C}$, so that

$$\hat{\hat{C}} = \int_{-\infty}^{\infty}\hat{C}\exp(ikx)\,dx = \sum_{n=0}^{\infty}\frac{(ik)^n}{n!}\int_{-\infty}^{\infty}x^n\hat{C}\,dx = \sum_{n=0}^{\infty}\frac{(ik)^n}{n!}\hat{C}_n, \quad \text{(B 2)}$$

say. It is also convenient to define

$$\mathscr{C}_n^{(0)} = \int_{-\infty}^{\infty}x^n\mathscr{C}^{(0)}\,dx. \quad \text{(B 3)}$$

On taking the Fourier transform of (B 1), and equating coefficients of $(ik)^n$, an equation for $\hat{C}_n$ is obtained, which is the Laplace transform of the equation for C_n obtained by Aris (1956). Aris showed that, as $t\to\infty$,

$$C_n \propto t^{\frac{1}{2}n}, \text{ if } n \text{ is even,} \quad \text{and} \quad C_n \propto t^{\frac{1}{2}(n-1)}, \text{ if } n \text{ is odd,}$$

so that $\hat{C}_n$ has the following development near $p = 0$:

$$\bar{C}_n \sim \begin{cases} \dfrac{f^{(n)}_{-(1+m)}}{p^{1+m}}+\dfrac{f^{(n)}_{-m}}{p^m}+\ldots+\dfrac{f^{(n)}_{-1}}{p}+f_0^{(n)}+\ldots, & \text{if} \quad n=2m, \\ \dfrac{f^{(n)}_{-(1+m)}}{p^{1+m}}+\dfrac{f^{(n)}_{-m}}{p^m}+\ldots+\dfrac{f^{(n)}_{-1}}{p}+f_0^{(n)}+\ldots, & \text{if} \quad n=2m+1. \end{cases} \quad \text{(B 4)}$$

The equation for $\hat{C}_0$ is

$$\frac{\partial}{\partial Y}\left(K\frac{\partial \hat{C}_0}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial \hat{C}_0}{\partial Z}\right)-\frac{pa^2}{D}\hat{C}_0 = -\frac{a^2}{D}\mathscr{C}_0^{(0)}. \quad \text{(B 5)}$$

Now, from (B 4),
$$\hat{C}_0 \sim \frac{f^{(0)}_{-1}}{p}+f_0^{(0)}+\ldots.$$

When this is substituted into (B 5) and coefficients of p^{-r} are compared, equations are obtained for $f_r^{(0)}$. However, the solution of (B 5) is particularly simple for the special case when $\mathscr{C}^{(0)}$ (and so each $\mathscr{C}_n^{(0)}$) is independent of Y and Z. This means the initial distribution of C is uniform over the pipe cross-section. Under these conditions,

$$\hat{C}_0 = \mathscr{C}_0^{(0)}/p. \quad \text{(B 6)}$$

The equation for $\hat{C}_1$ is, on using (B 6),

$$\frac{\partial}{\partial Y}\left(K\frac{\partial \hat{C}_1}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial \hat{C}_1}{\partial Z}\right)-\frac{pa^2}{D}\hat{C}_1 = -\frac{a^2}{D}\mathscr{C}_1^{(0)}-\frac{Ua^2}{D}\frac{V\mathscr{C}_0^{(0)}}{p}. \quad \text{(B 7)}$$

Now, from (B 4),

$$\hat{C}_1 \sim \frac{f^{(1)}_{-1}}{p}+f_0^{(1)}+f_1^{(1)}p+\ldots.$$

Thus, equating the coefficient of p^{-1} in (B 7) to zero,

$$\frac{\partial}{\partial Y}\left(K\frac{\partial f_{-1}^{(1)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial f_{-1}^{(1)}}{\partial Z}\right) = -\frac{Ua^2}{D}V\mathscr{C}_0^{(0)},$$

with solution, satisfying the zero normal gradient boundary condition,

$$f_{-1}^{(1)} = \text{const.} - \frac{Ua^2}{D}\mathscr{C}_0^{(0)}g^{(1)}.$$

The coefficient of p^0 in (B 7) gives the equation,

$$\frac{\partial}{\partial Y}\left(K\frac{\partial f_{0}^{(1)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial f_{0}^{(1)}}{\partial Z}\right) = \frac{a^2}{D}(f_{-1}^{(1)}-\mathscr{C}_1^{(0)});$$

and the right-hand side of this equation must have zero mean, for the reason discussed in §2. Hence, $\overline{f_{-1}^{(1)}} = \mathscr{C}_1^{(0)}$, so that the constant in the expression for $f_{-1}^{(1)}$ is $\mathscr{C}_1^{(0)}$. If axes are chosen so that $\mathscr{C}_1^{(0)} = 0$, then

$$f_{-1}^{(1)} = -\frac{Ua^2}{D}\mathscr{C}_0^{(0)}g^{(1)}, \tag{B 8}$$

and the equation for $f_0^{(1)}$ reduces to

$$\frac{\partial}{\partial Y}\left(K\frac{\partial f_{0}^{(1)}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial f_{0}^{(1)}}{\partial Z}\right) = -\frac{Ua^4}{D^2}\mathscr{C}_0^{(0)}g^{(1)}.$$

The solution of this equation can be written symbolically

$$f_0^{(1)} = \text{const.} - \frac{Ua^4}{D^2}\mathscr{C}_0^{(0)}\{Ig^{(1)}\},$$

where $\{Ig^{(1)}\}$ satisfies

$$\overline{\{Ig^{(1)}\}} = 0, \quad \text{and} \quad \frac{\partial}{\partial Y}\left(K\frac{\partial\{Ig^{(1)}\}}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial\{Ig^{(1)}\}}{\partial Z}\right) = g^{(1)}.$$

The constant in this expression must be zero if the equation for $f_1^{(1)}$ is soluble. Hence,

$$f_0^{(1)} = -\left(\frac{Ua^2}{D}\right)\left(\frac{a^2}{D}\right)\mathscr{C}_0^{(0)}\{Ig^{(1)}\}. \tag{B 9}$$

Similarly,

$$f_1^{(1)} = -\left(\frac{Ua^2}{D}\right)\left(\frac{a^2}{D}\right)^2\mathscr{C}_0^{(0)}\{I^2g^{(1)}\}, \tag{B 10}$$

where $\{I^2g^{(1)}\}$ is related to $\{Ig^{(1)}\}$ in the same way as $\{Ig^{(1)}\}$ is related to $g^{(1)}$. Since $\overline{g^{(1)}} = \overline{\{Ig^{(1)}\}} = \overline{\{I^2g^{(1)}\}} = 0$, it follows, from (B 8), (B 9) and (B 10), that

$$\overline{\hat{C}_1} = 0,$$

since $\mathscr{C}_1^{(0)} = 0$. Hence, for all t,

$$\overline{C_1} = \int_{-\infty}^{\infty} x\overline{C}\,dx = 0 \Rightarrow x_g = 0. \tag{B 11}$$

The equation for $\hat{C}_2$ is found to be

$$\frac{\partial}{\partial Y}\left(K\frac{\partial \hat{C}_2}{\partial Y}\right)+\frac{\partial}{\partial Z}\left(K\frac{\partial \hat{C}_2}{\partial Z}\right)-\frac{pa^2}{D}\hat{C}_2 = -\frac{a^2}{D}\mathscr{C}_2^{(0)}-\frac{2Ka^2}{p}\mathscr{C}_0^{(0)}-\frac{2Ua^2}{D}V\hat{C}_1. \tag{B 12}$$

The values of the functions appearing in the expansion of $\hat{C}_2$ about $p = 0$ can be found in the way described above for those connected with $\hat{C}_1$. The results are

$$\left.\begin{aligned} f_{-2}^{(2)} &= 2\mathscr{C}_0^{(0)}\left\{D - \frac{U^2a^2}{D}\,\overline{Vg^{(1)}}\right\} = M\mathscr{C}_0^{(0)}\frac{U^2a^2}{D}, \\ f_{-1}^{(2)} &= \mathscr{C}_2^{(0)} + 2\mathscr{C}_0^{(0)}\left(\frac{Ua^2}{D}\right)^2 \{g^{(2)} - \overline{V\{Ig^{(1)}\}}\}, \\ f_0^{(2)} &= 2\mathscr{C}_0^{(0)}\left(\frac{Ua^2}{D}\right)^2\left(\frac{a^2}{D}\right)[\{Ig^{(2)}\} + \{I(V\{Ig^{(1)}\} - \overline{V\{Ig^{(1)}\}})\}]. \end{aligned}\right\} \tag{B 13}$$

Hence, on taking the mean over the cross-section,

$$\overline{\hat{C}}_2 \sim \frac{MU^2a^2\mathscr{C}_0^{(0)}}{D}\frac{}{p^2} + \frac{\{\mathscr{C}_2^{(0)} - 2\mathscr{C}_0^{(0)}(Ua^2/D)^2\,\overline{V\{Ig^{(1)}\}}\}}{p} + \ldots,$$

so that, as $t \to \infty$,

$$\nu_2(t) = \frac{\overline{C}_2}{\mathscr{C}_0^{(0)}} \sim \frac{MU^2a^2t}{D} + \left\{\nu_2(0) - 2\left(\frac{Ua^2}{D}\right)^2 \overline{V\{Ig^{(1)}\}}\right\}. \tag{B 14}$$

However, since

$$\frac{\partial}{\partial Y}\left(K\frac{\partial\{Ig^{(1)}\}}{\partial Y}\right) + \frac{\partial}{\partial Z}\left(K\frac{\partial\{Ig^{(1)}\}}{\partial Z}\right) = g^{(1)},$$

$$\begin{aligned} \overline{\{g^{(1)}\}^2} &= \overline{g^{(1)}[\nabla\,.\,K\nabla\{Ig^{(1)}\}]}, \quad \text{where} \quad \nabla \equiv \left(\frac{\partial}{\partial Y}, \frac{\partial}{\partial Z}\right). \\ &= -\overline{K\nabla g^{(1)}\,.\,\nabla\{Ig^{(1)}\}} = \overline{\{Ig^{(1)}\}\nabla\,.\,(K\nabla g^{(1)})} \\ &= \overline{V\{Ig^{(1)}\}}. \end{aligned}$$

on using the defining equation of $g^{(1)}$, (1.10). Thus, substituting in (B 14),

$$\nu_2(t) \sim \frac{MU^2a^2t}{D} + \left\{\nu_2(0) - 2\left(\frac{Ua^2}{D}\right)^2 \overline{\{g^{(1)}\}^2}\right\}, \tag{B 15}$$

which is just (3.8).

The equation for $\hat{C}_3$ is

$$\frac{\partial}{\partial Y}\left(K\frac{\partial\hat{C}_3}{\partial Y}\right) + \frac{\partial}{\partial Z}\left(K\frac{\partial\hat{C}_3}{\partial Z}\right) - \frac{pa^2}{D}\hat{C}_3 = -\frac{a^2}{D}\mathscr{C}_3^{(0)} - 6Ka^2\hat{C}_1 - 3\left(\frac{Ua^2}{D}\right)V\hat{C}_2. \tag{B 16}$$

The value of $\overline{\hat{C}_3}$ can be found by taking the mean of this equation, using the results above for the terms in $\hat{C}_1$ and $\hat{C}_2$. On taking the Laplace transform of this value, it is found that, as $t \to \infty$,

$$\begin{aligned} \nu_3(t) &= \overline{C_3}/\mathscr{C}_0^{(0)} \\ &\sim 6\left(\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}\right)\left(\frac{MU^2a^2t}{D}\right)\left(\frac{Ua^2}{D}\right) \\ &\quad + \left\{\nu_3(0) + 6\left(\frac{Ua^2}{D}\right)^3\left[\overline{V\{Ig^{(2)}\}} + \overline{V\{I(V\{Ig^{(1)}\} - \overline{V\{Ig^{(1)}\}})\}} - \left(\frac{D}{Ua}\right)^2 \overline{K\{Ig^{(1)}\}}\right]\right\}. \end{aligned} \tag{B 17}$$

The constant term in this expression can be simplified by the same processes that were used in going from (B 14) to (B 15). It is easily shown that

$$\overline{V\{Ig^{(2)}\}} = \overline{g^{(1)}g^{(2)}},$$

and that
$$\overline{V\{I(V\{Ig^{(1)}\} - \overline{V\{Ig^{(1)}\}})\}} - \left(\frac{D}{Ua}\right)^2 \overline{K\{Ig^{(1)}\}} = \overline{g^{(1)}g^{(2)}}.$$

Thus,
$$\nu_3(t) \sim 6\left(\frac{\alpha^{(1,1)}}{\alpha^{(0,0)}}\right)\left(\frac{MU^2a^2t}{D}\right)\left(\frac{Ua^2}{D}\right) + \left\{\nu_3(0) + 12\left(\frac{Ua^2}{D}\right)^3 \overline{g^{(1)}g^{(2)}}\right\}, \tag{B 18}$$

which is just (3.9).

Appendix C. The exact values of $\nu_2(t)$ and $\nu_3(t)$ in Poiseuille flow

This appendix shows how $\nu_2(t)$ and $\nu_3(t)$ can be obtained exactly for Poiseuille flow in which, with $R = (Y^2+Z^2)^{\frac{1}{2}}$,

$$V(Y,Z) = 1 - 2R^2 \quad \text{and} \quad K(Y,Z) = 1.$$

The only case that is considered is that when

$$\mathscr{C}^{(0)}(X, Y, Z) \propto \delta(X), \tag{C 1}$$

so that $\nu_n(0) = 0$, and the method adopted is to solve the appropriate moment equations of Aris. These can be solved in the manner described by Aris (1956), but the method chosen here is to solve the Laplace transforms of the moment equations. This method gives the results (4.9) and (4.10) directly.

The equation (B 7) for $\hat{C}_1$, with the values of V and K given above, is

$$\frac{d^2\hat{C}_1}{dR^2} + \frac{1}{R}\frac{d\hat{C}_1}{dR} - \frac{pa^2}{D}\hat{C}_1 = -\left(\frac{Ua^2}{D}\right)\frac{V\mathscr{C}_0^{(0)}}{p}, \tag{C 2}$$

since $\mathscr{C}_1^{(0)} = 0$. The solution of this equation, satisfying

$$d\hat{C}_1/dR = 0 \quad \text{at} \quad R = 1,$$

is easily verified to be

$$\hat{C}_1 = \left(\frac{Ua^2}{D}\right)\left(\frac{\mathscr{C}_0^{(0)}}{p}\right)\left(\frac{D}{pa^2}\right)\left[(1-2R^2) - 8\left(\frac{D}{pa^2}\right) + \frac{4}{q}\frac{I_0(qR)}{I_1(q)}\right], \tag{C 3}$$

where
$$q = (pa^2/D)^{\frac{1}{2}}, \tag{C 4}$$

and I_0 and I_1 are modified Bessel functions.

The equation (B 12) for $\hat{C}_2$ is

$$\frac{d^2\hat{C}_2}{dR^2} + \frac{1}{R}\frac{d\hat{C}_2}{dR} - \frac{pa^2}{D}\hat{C}_2 = -2\left(\frac{Ua^2}{D}\right)V\hat{C}_1 - \frac{2a^2}{p}\mathscr{C}_0^{(0)},$$

and the solution of this equation, satisfying $d\hat{C}_2/dR = 0$ at $R = 1$, can be verified to be

$$\begin{aligned}\hat{C}_2 = \frac{2D\mathscr{C}_0^{(0)}}{p^2} + 2\left(\frac{Ua^2}{D}\right)^2\frac{\mathscr{C}_0^{(0)}}{p}\left(\frac{D}{pa^2}\right)\Bigg[&\left\{\left(\frac{1}{q^2} - \frac{24}{q^4} + \frac{320}{q^6}\right) - \left(\frac{4}{q^2} - \frac{80}{q^4}\right)R^2 + \frac{4}{q^2}R^4\right\} \\ &- \frac{4}{qI_1(q)}\left\{\left(\frac{1}{2q} - \frac{2}{3q^3}\right)RI_1(qR) - \frac{1}{3q}R^3I_1(qR) + \frac{1}{3q^2}R^2I_0(qR)\right\} \\ &- \left\{\frac{28}{3q^3I_1(q)} + \frac{160}{q^5I_1(q)} - \frac{2I_0(q)}{3q^2I_1^2(q)}\right\}I_0(qR)\Bigg].\end{aligned} \tag{C 5}$$

The mean of this equation over the cross-section gives

$$\overline{\hat{C}_2} = \frac{2D\mathscr{C}_0^{(0)}}{p^2} + 4\left(\frac{Ua^2}{D}\right)^2 \frac{\mathscr{C}_0^{(0)}}{p}\left(\frac{D}{pa^2}\right)^2\left[\frac{1}{6} - \frac{4}{q^2} - \frac{32}{q^4} + \frac{16I_0(q)}{q^3 I_1(q)}\right],$$

and it may be shown that the inverse Laplace transform of this is (4.9).

The equation for $\hat{C}_3$ is given in (B 16). On taking the cross-sectional mean of this equation, it is found that

$$p\overline{\hat{C}_3} = 3U\overline{V\hat{C}_2},$$

since $\overline{\hat{C}_1} = 0$. On multiplying (C 5) by V, and integrating, it is found that

$$\overline{\hat{C}_3} = 32\left(\frac{Ua^2}{D}\right)^3 \frac{\mathscr{C}_0^{(0)}}{p}\left(\frac{D}{pa^2}\right)^2 \times\left[\frac{(q^4+172q^2+960)\,I_1^2(q) - (28q^3+480q)\,I_0(q)\,I_1(q) + q^4 I_0^2(q)}{q^8 I_1^2(q)}\right], \quad \text{(C 6)}$$

and the inverse Laplace transform of this is (4.10).

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