J. Fluid Mech. (1967), *vol.* 29, *part* 3, *pp.* 465–484
Printed in Great Britain

The effects of curvature and buoyancy on the laminar dispersion of solute in a horizontal tube

By M. EMIN ERDOGAN† AND P. C. CHATWIN
Department of Applied Mathematics and Theoretical Physics,
University of Cambridge

(Received 31 October 1966)

When a solute is injected into a straight circular tube through which a solvent is in steady laminar flow it spreads out longitudinally under the combined effect of molecular diffusion and advection with the flow. Taylor (1953) showed that, provided there is no density difference between the solute and the solvent, the distribution of mean concentration satisfies a diffusion equation with a certain longitudinal diffusivity for large times and with respect to axes moving with the discharge velocity. In the present paper it is shown by a general argument that this statement remains true if the pipe is uniformly curved. An expression is given for the diffusivity, valid when the radius of curvature is sufficiently large. For all common liquids and most gases the diffusivity is reduced by the curvature. The rest of the paper deals with the effects of buoyancy forces caused by a density difference between the solute and the solvent, when the tube is horizontal. It is shown that in general a longitudinal diffusivity does not exist and an equation is derived that replaces the diffusion equation if the effects of buoyancy are small. A prediction from this equation is that buoyancy should not have a noticeable effect on the longitudinal dispersion for Péclet numbers near a certain value at which the two opposing influences of horizontal spreading due to gravity and lateral mixing due to secondary flow are in balance. This prediction is consistent with some observations made by Reejhsinghani, Gill & Barduhn (1966).

1. Introduction

This paper is concerned with the longitudinal dispersion of a solute, subject to molecular diffusion, when it is introduced into a circular tube through which a solvent is in steady laminar flow. It was first shown by Taylor (1953) that the combined effect of molecular diffusion in a lateral plane and of advection on the distribution of concentration of a cloud of solute is ultimately to make it spread out symmetrically about a point moving with the mean flow velocity W. The mean concentration over the cross-section satisfies a diffusion equation asymptotically (i.e. as $t \to \infty$), with a certain longitudinal diffusivity. For the case of Poiseuille flow in a straight circular tube, and with neglect of molecular diffusion in the axial direction, this diffusivity is $a^2 W^2/48\kappa$ where κ is the molecular diffusivity and a the pipe radius. It was later shown by Aris (1956) that axial mole-

---

† Present address: Division of Mechanics and Fluid Mechanics, Technical University of Istanbul, Istanbul, Turkey.

cular diffusion makes an independent contribution so that the total longitudinal diffusivity D is

$$D = \kappa + \frac{a^2 W^2}{48\kappa}. \tag{1.1}$$

Numerical work by Ananthakrishnan, Gill & Barduhn (1965) shows that Taylor's theory, without Aris's modification, is valid provided that the Péclet number $P(= Wa/\kappa)$ is greater than 50 and the time after injection is greater than a^2/κ, and that with Aris's modification it is asymptotically valid for all P.

The applications of this theory appear numerous. Taylor originally suggested that values of κ might be found conveniently by measuring D. Amongst other applications are those to chromatographic columns (Van Deemter, Broeder & Lauwerier 1956), distillation processes (Aris 1959) and blood flow (see the discussion in Lighthill 1966).

In the present paper we discuss the applications of Taylor's theory to dispersion of a neutrally buoyant solute in a circular pipe curved in a circular arc, and to the dispersion of a non-neutrally buoyant solute in a straight horizontal circular pipe. It is convenient, first of all, to recall the arguments used by Batchelor (1966) which give a definite sufficient criterion for the existence of a longitudinal diffusivity. These arguments were originally presented in the context of the probability distribution of a single fluid particle in turbulent flow through a pipe, but, as Batchelor points out, they are widely applicable. It is known that if the velocity of a fluid particle in a particular direction is a stationary random function of time, the dispersion $\{\overline{(Y-\overline{Y})^2}\}^{\frac{1}{2}}$ increases as $t^{\frac{1}{2}}$, as $t \to \infty$, where t is the time that has elapsed since the particle's position was known with certainty. Here the overbars denote ensemble means, and $Y = Y(t)$ is the displacement of the fluid particle in the direction under consideration. Also the probability distribution of $Y(t)$ tends to a Gaussian form as $t \to \infty$. If now we regard the solute as a passive marker of the fluid molecules, these arguments are directly applicable to the case of the longitudinal dispersion of a cloud of solute, subject to molecular diffusion, which is advected by a solvent in laminar flow through a tube (or subject to turbulent diffusion in addition, if the flow is turbulent). All that is required is that flow conditions be uniform in the longitudinal direction, since the random phenomena called molecular diffusion and turbulent diffusion then have statistical properties which do not change with position down the tube. (In the case of molecular diffusion this condition is satisfied only if the molecular diffusion coefficient is independent of concentration. Throughout this paper it will be assumed that κ is independent of concentration.)

It should be pointed out that the concentration distribution need not have a Gaussian form asymptotically, although this will be so whenever the cloud of solute is initially of finite extent. The two statements, that as $t \to \infty$, (i) the probability distribution function of the displacement of a single fluid particle is a Gaussian function of Y, and (ii) the dispersion is proportional to $t^{\frac{1}{2}}$, are sufficient to show that the probability distribution function of a single fluid particle satisfies a diffusion equation as $t \to \infty$ with a constant diffusivity, with respect to axes chosen so that $\overline{Y} = 0$. It is then possible to deduce (Batchelor & Townsend 1956) that the distribution of concentration of a cloud of solute satisfies the same diffusion equation as $t \rightarrow \infty$ relative to axes moving with the mean velocity W. The asymptotic form of the distribution of concentration is then given by the relevant solution of this diffusion equation.

It now becomes obvious that in the first case mentioned above, that of flow in a uniformly curved pipe, there will exist a longitudinal diffusivity. The secondary flow induced by the curvature has properties which do not change in the direction of the pipe axis, and this is the only necessary condition. The components of this secondary flow in the cross-sectional plane increase the lateral mixing, and make the distribution of concentration in a cross-sectional plane more uniform so that the flow is less effective in dispersing the material longitudinally. It may therefore be expected that the longitudinal diffusivity is reduced by the curvature, at any rate for small curvature when the secondary flow may be regarded as a perturbation of the basic Poiseuille flow. A lowering of the dispersion has been observed by Evans & Kenney (1965), and by Caro (1966). It is interesting to note that Taylor (1954*a*) reported some experiments on turbulent flow in a curved channel, and observed a large increase in the longitudinal diffusivity. This is difficult to explain on the above argument, which should hold even in a turbulent flow, but Taylor (1966), in a private communication, has pointed out that the intensity of the turbulence is probably much reduced in a curved pipe, relative to a straight pipe at the same Reynolds number. This lowering of intensity will act to increase the dispersion since the turbulent diffusivity is lowered; presumably in Taylor's experiments this effect is much greater than that due to the secondary flow.

In this paper an expression is given for the change in D in laminar flow due to the curvature, when this curvature is sufficiently small for the expressions for the secondary flow given by Dean (1927, 1928) to be valid. For all common liquids and for most gases, this expression does indeed predict a lowering of D.

When one comes to the case in which the pipe is straight and horizontal, but the solute is of a different density from the ambient fluid, the problem is more complicated. Buoyancy forces associated with the density gradients set up secondary flows, and now the conditions are not stationary in the axial direction in general. The solute no longer acts as a passive marker of fluid particles since it gives rise to definite dynamical effects, and it is only in one special case, corresponding to a uniform axial concentration gradient, that these effects are uniform in the longitudinal direction.

In work by Reejhsinghani *et al.* (1966) some observations are reported on this problem. Their experiments were carried out with duPont's 'Pontamine' 6BX dye in distilled water in two horizontal tubes, of 1·5 and 5 mm. internal diameter. The maximum density difference between the dye solution and the pure water was $1{\cdot}2 \times 10^{-4}$ g/ml. For P below a certain critical value P_c (which presumably depends on other variables like the ratio ν/κ, but was approximately 22·5 for the 1·5 mm. tube) the dispersion was much greater than that predicted by the Taylor–Aris formula, but for values of P greater than P_c the dispersion was less. A qualitative explanation of these observations was given. There are two effects of the density difference. First, the axial density gradient causes an axial pressure gradient, and so a change in the axial velocity distribution, which, acting by itself, may increase the dispersion. A calculation of this effect was made by Taylor in 1953 (but not published) in connexion with his original experiments. The second effect is due to the secondary flow in the cross-sectional plane set up by the radial variation of density. As in the case of the curved pipe this would be expected to decrease the dispersion. To explain the dependence on P, Reejhsinghani *et al.* refer to the results of a paper by del Casal & Gill (1962) on the similar problem of the effects of buoyancy caused by a uniform axial temperature gradient on flow in a horizontal tube (but it must be noted that the boundary conditions for concentration are not the same as those used for temperature in the above paper so the analogy is not exact) in which the change in the friction factor caused by the above two effects was evaluated. Del Casal & Gill found that as P increased the effect on the friction factor of the secondary flow became more important than that due to the axial velocity change. Reejhsinghani *et al.* then inferred that the change in the dispersion was also dominated by the secondary flow for large P and thus should be negative in agreement with their observations.

In the present paper some calculations are presented which show the interaction of the above effects and which are compared with the observations of Reejhsinghani *et al.* However, full understanding of the situation is not claimed. This must await a more detailed treatment (theoretical or numerical) of the full non-steady equations.

2. Laminar dispersion in a straight circular pipe with no buoyancy

In this section a summary of the original work of Taylor and Aris is given for later use. We take axes as shown in figure 1. The equation for the concentration distribution C is

$$\frac{\partial C}{\partial t}+W(1-2r^2)\frac{\partial C}{\partial X}=\frac{\kappa}{a^2}\left[\frac{\partial^2 C}{\partial r^2}+\frac{1}{r}\frac{\partial C}{\partial r}+a^2\frac{\partial^2 C}{\partial X^2}\right], \tag{2.1}$$

assuming that there is rotational symmetry of C. Here the axes are moving with the mean velocity so that $X = x - Wt$.

The boundary conditions are:

$$\partial C/\partial r = 0 \text{ at } r = 1; \quad C \text{ finite at } r = 0. \tag{2.2}$$

In his original analysis Taylor made the following assumptions:

(i) the transfer of C in the axial direction by molecular diffusion is small compared with that caused by advection, so that $a^2\,\partial^2 C/\partial X^2$ is assumed negligible;

(ii) relative to axes moving with the mean velocity the motion is approximately steady so that $\partial C/\partial t = 0$; Taylor showed that this means that the time-scale necessary for appreciable effects due to advection to appear (which is of the order of L/W, where L is the length of pipe over which the material is spread) is much greater than that in which radial variations of concentration die away under the action of molecular diffusion; this is of the order of a^2/κ so that the condition is $L \gg Wa^2/\kappa$ (Taylor 1954*b*).

With these assumptions (2.1) becomes

$$W(1-2r^2)\frac{\partial C}{\partial X}=\frac{\kappa}{a^2}\left(\frac{\partial^2 C}{\partial r^2}+\frac{1}{r}\frac{\partial C}{\partial r}\right), \tag{2.3}$$

and the solution is

$$C=\beta X+\beta\frac{Wa^2}{8\kappa}(2r^2-r^4), \tag{2.4}$$

where β is a constant. The mean concentration over a cross-section is C_m, where

$$\begin{aligned} C_m &= 2\int_0^1 Cr\,dr \\ &= \beta X + \beta\frac{Wa^2}{12\kappa}, \end{aligned} \tag{2.5}$$

so that

$$C = C_m + \frac{\partial C_m}{\partial X}\frac{Wa^2}{24\kappa}(-2+6r^2-3r^4). \tag{2.6}$$

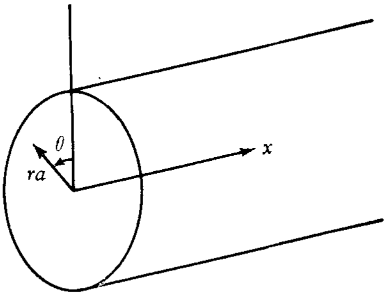

FIGURE 1. System of co-ordinates for straight pipe; $(ra,\ \theta,\ x)$ are cylindrical polars and $\theta = 0$ is the upward vertical.

Now the rate of transfer of concentration across a plane moving with the mean velocity depends only on the radial distribution of C, and is

$$\begin{aligned} Q &= 2\pi a^2\int_0^1 W(1-2r^2)Cr\,dr \\ &= -\pi a^2\left(\frac{a^2W^2}{48\kappa}\right)\left(\frac{\partial C_m}{\partial X}\right). \end{aligned} \tag{2.7}$$

As Taylor pointed out, this expression has exactly the same form as if C_m were being diffused across planes moving with a velocity W by a process which obeys the diffusion law, with a diffusivity $a^2W^2/48\kappa$.

Taylor now assumed that the same relationship between Q and $\partial C_m/\partial X$ holds approximately, even if $\partial C/\partial X$ and $\partial C/\partial t$ are not constant, and thus that C_m satisfies a (one-dimensional) diffusion equation with diffusivity $a^2W^2/48\kappa$.

Aris showed, by taking integral moments of (2.1), that, when the effect of axial molecular diffusion is included, this conclusion remains true but with a total longitudinal diffusivity given by

$$D = \kappa + a^2W^2/48\kappa. \tag{2.8}$$

Taylor's result is a good approximation for large P.

Another proof of (2.8) is given here for later use. Taking the average of (2.1) over the cross-section and using the boundary conditions yields

$$\frac{\partial C_m}{\partial t} = \kappa\frac{\partial^2 C_m}{\partial X^2} - \frac{\partial}{\partial X}\{wC\}_m, \tag{2.9}$$

where $w = W(1-2r^2)$. Now $\{wC\}_m$ represents the total advective flux across a plane moving with velocity W, and we know from the arguments in § 1 that this is of the same form as would be given by a diffusion process as $t \to \infty$, that is, it is given by

$$\{wC\}_m = -D' \frac{\partial C_m}{\partial X}, \tag{2.10}$$

where D' is a constant. We are then able to find D' by choosing C_m to be exactly linear (Batchelor 1966). Thus we follow Taylor's original analysis, and obtain $D' = a^2 W^2/48\kappa$ so that (2.9) becomes, as $t \to \infty$,

$$\begin{aligned}\frac{\partial C_m}{\partial t} &= (\kappa + D') \frac{\partial^2 C_m}{\partial X^2} \\ &= \left(\kappa + \frac{a^2 W^2}{48\kappa}\right) \frac{\partial^2 C_m}{\partial X^2},\end{aligned} \tag{2.11}$$

agreeing with (2.8).

Both Taylor's analysis and Aris's extension are in agreement with numerical work and with experimental observations. The only reported disagreement is that given by Bournia, Coull & Houghton (1961), but this is almost certainly due to density effects (see the discussion in Reejhsinghani *et al.* (1966)).

3. Laminar dispersion in a curved circular pipe with no buoyancy

The velocity profiles

The system of co-ordinates used in this section is shown in figure 2. *ZOC* and *BOC* are a vertical and a horizontal plane respectively. *O* is the centre of curvature, and *b* the radius of curvature, of the coil in which the pipe is wound. It will be assumed that the motion is steady and independent of ϕ, except for the forcing pressure, which is a linear function of ϕ. In fact we shall write

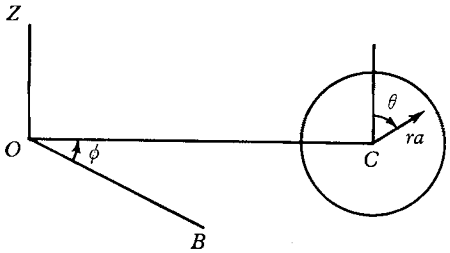

FIGURE 2. System of co-ordinates for curved pipe.

$$-\frac{1}{b}\frac{\partial}{\partial \phi}\left(\frac{p'}{\rho_0}\right) = \frac{8\nu W_0}{a^2}, \tag{3.1}$$

where p' is the pressure and ρ_0 the fluid density: note that W_0 is then the mean velocity if the pipe is straight (when $(1/b)\,\partial/\partial\phi$ is replaced by $\partial/\partial x$, where x is in the axial direction). We then non-dimensionalize the full equations of motion as follows. The length-scale for the radial co-ordinate is a (as already indicated in figure 2), the velocity scale for the axial velocity is W_0, that for the cross-sectional velocities is ν/a, and the pressure variation in a cross-sectional plane is non-dimensionalized by $\rho_0\nu^2/a^2$. The full equations of motion are then (Dean 1927):

$$\left.\begin{aligned}
&u\frac{\partial u}{\partial r}+\frac{v}{r}\frac{\partial u}{\partial \theta}-\frac{v^2}{r}-N\frac{w^2\sin\theta}{\delta} = -\frac{\partial p}{\partial r}-\left(\frac{1}{r}\frac{\partial}{\partial \theta}+\frac{\epsilon\cos\theta}{\delta}\right)\left(\frac{\partial v}{\partial r}+\frac{v}{r}-\frac{1}{r}\frac{\partial u}{\partial \theta}\right),\\
&u\frac{\partial v}{\partial r}+\frac{v}{r}\frac{\partial v}{\partial \theta}+\frac{uv}{r}-N\frac{w^2\cos\theta}{\delta} = -\frac{1}{r}\frac{\partial p}{\partial \theta}+\left(\frac{\partial}{\partial r}+\frac{\epsilon\sin\theta}{\delta}\right)\left(\frac{\partial v}{\partial r}+\frac{v}{r}-\frac{1}{r}\frac{\partial u}{\partial \theta}\right),\\
&u\frac{\partial w}{\partial r}+\frac{v}{r}\frac{\partial w}{\partial \theta}+\frac{\epsilon uw\sin\theta}{\delta}+\frac{\epsilon vw\cos\theta}{\delta}\\
&\qquad = \frac{8}{\delta}+\left[\left(\frac{\partial}{\partial r}+\frac{1}{r}\right)\left(\frac{\partial w}{\partial r}+\frac{\epsilon w\sin\theta}{\delta}\right)+\frac{1}{r}\frac{\partial}{\partial \theta}\left(\frac{1}{r}\frac{\partial w}{\partial \theta}+\frac{\epsilon w\cos\theta}{\delta}\right)\right].
\end{aligned}\right\} \tag{3.2}$$

Here (u, v, w) are the non-dimensional velocity components corresponding to (r, θ, ϕ) respectively and

$$p = \frac{a^2}{\rho_0\nu^2}\left[p'-p'_0+\frac{8\nu b W_0\rho_0}{a^2}\phi\right], \tag{3.3}$$

where p'_0 is a reference pressure and p is independent of ϕ. The equation of continuity can be satisfied identically by writing

$$u = \frac{1}{r\delta}\frac{\partial \Psi}{\partial \theta}, \quad v = -\frac{1}{\delta}\frac{\partial \Psi}{\partial r}. \tag{3.4}$$

We have written

$$\epsilon = \frac{a}{b}; \quad N = \frac{a}{b}\left(\frac{W_0 a}{\nu}\right)^2; \quad \delta = 1+\epsilon r\sin\theta. \tag{3.5}$$

Thus there are two independent parameters, ϵ and N, in the problem. ϵ specifies the geometry of the system and N the dynamics. If ϵ (and so N, provided $W_0 a/\nu$ is finite) is zero the solution (with a suitable choice for the zero of Ψ) is:

$$\Psi = 0; \quad w = 2(1-r^2).$$

It will be assumed that Ψ and $w-2(1-r^2)$ can be written as double power series in ϵ and N, the general term of which (for Ψ) is $\Psi_{mn}\epsilon^m N^n$.

In principle the terms in these series may be found by eliminating p from the first two equations of (3.2), substituting in the series expansions and comparing coefficients. Since $r = 1$ is a streamline Ψ_{m0} will be zero for all m; despite this fact the procedure will become very heavy after one or two terms. In this paper it will be assumed that there are ranges of values of ϵ and N for which the following forms, given by Dean (1928) and Cuming (1952), are good approximations:

$$\Psi = N\Psi_{01}+N^2\Psi_{02}; \quad w-2(1-r^2) = Nw_{01}+N^2w_{02}. \tag{3.6}$$

The main ideas and arguments behind this rather drastic approximation can be summarized as follows.

(*a*) Dean (1928) pointed out that taking the limit $\epsilon \to 0$ in the equations while keeping N fixed retains the main dynamical effects due to centrifugal acceleration, but eliminates purely geometrical effects.

(*b*) If there is a range of ϵ for which (*a*) is valid, N should be a dynamical similarity coefficient for the flow. That this is true for laminar flow was verified by White

(1929), who plotted the ratio of the friction coefficient in a curved pipe to that in a straight pipe at the same Reynolds number against N, and found that the results of four sets of experiments with values of ϵ ranging from 0·0664 to 0·0005 lay on a single curve (but ϵ was important in determining the critical Reynolds number for the onset of turbulence).

(c) Cuming (1952) extended Dean's work to cover the case when the pipe is of elliptical cross-section, and gave an expression for w which is the first two terms of the full expansion. He made the remark that in the circular case ϵw_{10} is numerically small compared with $N w_{01}$; because of this he neglected w_{10}, w_{20} and w_{11} when calculating the second-order terms. This reasoning is difficult to follow since the relative sizes of ϵ and N must be relevant, but Truesdell (1963) found that his exact numerical solution for $\epsilon = 0{\cdot}01$ and $N = 0{\cdot}25$ agreed very well with Cuming's approximate solution.

These arguments suggest that if ϵ is low enough its neglect is justifiable. The series expansions then reduce to terms in N alone. However the cutting off of these series after the N^2-terms will place an upper limit on N. Dean suggests that this upper limit is about 50 but there appears to be little or no relevant information about the value of this limit. If $\epsilon \rightarrow 0$ the equations (3.4) reduce to

$$u = \frac{1}{r}\frac{\partial \Psi}{\partial \theta}; \quad v = -\frac{\partial \Psi}{\partial r}. \tag{3.7,}$$

Dean (1928) gives the following expressions, which we shall use in our evaluation of the distribution of concentration:

$$\left.\begin{aligned} \Psi_{01} &= f_1(r)\cos\theta, \quad & \Psi_{02} &= f_2(r)\sin 2\theta; \\ w_{01} &= g_1(r)\sin\theta, \quad & w_{02} &= g_2(r) + \overline{g_2(r)}\cos 2\theta. \end{aligned}\right\} \tag{3.8}$$

For our purposes the values of $f_2(r)$ and $\overline{g_2(r)}$ are not needed. The forms of $f_1(r)$-$g_1(r)$ and $g_2(r)$ are given in an appendix.

The mean axial velocity is W, where

$$\begin{aligned} W &= \frac{1}{\pi}\int_0^{2\pi} d\theta \int_0^1 W_0 w r\, dr \\ &= W_0(1 - AN^2), \end{aligned} \tag{3.9}$$

where A is a constant whose value is given in the appendix.

The calculation of the concentration flux across a cross-section

The equation for C, the distribution of concentration, is non-dimensionalized using the same scales as previously. In addition, the time t is replaced by τ, where $\tau = \kappa t/a^2$ and the non-dimensional variable ξ is written for $(b\phi - Wt)/a$, so that the axes are moving with the mean velocity. Furthermore, it is again assumed that the limit $\epsilon \rightarrow 0$ can be taken provided ϵ is small. The equation obtained is

$$\frac{\partial C}{\partial \tau} - \frac{\sigma}{r}\frac{\partial(\Psi, C)}{\partial(r,\theta)} + \left(w\frac{W_0 a}{\kappa} - P\right)\frac{\partial C}{\partial \xi} = \frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{1}{r^2}\frac{\partial^2 C}{\partial \theta^2} + \frac{\partial^2 C}{\partial \xi^2}. \tag{3.10}$$

P is the Péclet number Wa/κ, and $\sigma = \nu/\kappa$.

It is known, for the reasons given in § 1, that a longitudinal diffusion coefficient exists asymptotically whatever the initial form of the concentration. It is also again true that the effect of axial molecular diffusion, that is of the term $\partial^2 C/\partial\xi^2$, on this diffusivity is simply additive. This can be shown exactly as in § 2, using the additional boundary condition that Ψ is constant at $r = 1$. It is thus convenient and simplest for the purpose of calculating this diffusivity to follow Taylor's original analysis, presented in § 2, and consider the case when $\partial C/\partial\xi$ is constant, since this distribution will be exactly steady in the axes we are using. Thus the equation to be solved is

$$\left(\frac{\partial^2 C}{\partial r^2}+\frac{1}{r}\frac{\partial C}{\partial r}+\frac{1}{r^2}\frac{\partial^2 C}{\partial \theta^2}\right)+\frac{\sigma}{r}\frac{\partial(\Psi, C)}{\partial(r,\theta)} = \left(\frac{W_0 a}{\kappa}w-P\right)\frac{\partial C}{\partial \xi}, \tag{3.11}$$

where $\partial C/\partial\xi$ is constant. The boundary conditions are given in equation (2.2).

We write

$$C-\xi\frac{\partial C_m}{\partial \xi} = \frac{\partial C_m}{\partial \xi}\frac{W_0 a}{\kappa}(C_0+NC_1+N^2C_2), \tag{3.12}$$

since $\partial C_m/\partial\xi = \partial C/\partial\xi$ when the latter is constant. Substituting in (3.11) and comparing coefficients, we find

$$\nabla^2 C_n = \begin{cases}(1-2r^2), & n=0;\\ w_{01}-\dfrac{\sigma}{r}\dfrac{\partial(\Psi_{01}, C_0)}{\partial(r,\theta)}, & n=1;\\ (w_{02}+A)-\dfrac{\sigma}{r}\dfrac{\partial(\Psi_{02}, C_0)}{\partial(r,\theta)}-\dfrac{\sigma}{r}\dfrac{\partial(\Psi_{01}, C_1)}{\partial(r,\theta)}, & n=2;\end{cases} \tag{3.13}$$

where

$$\nabla^2 \equiv \left(\frac{\partial^2}{\partial r^2}+\frac{1}{r}\frac{\partial}{\partial r}+\frac{1}{r^2}\frac{\partial^2}{\partial \theta^2}\right).$$

On using the expressions above for Ψ and w, the solutions are obtained in the form:

$$\left.\begin{aligned} C_0 &= \tfrac{1}{8}(2r^2-r^4),\\ C_1 &= h_1(r)\sin\theta,\\ C_2 &= h_2(r)+\overline{h_2(r)}\cos 2\theta.\end{aligned}\right\} \tag{3.14}$$

The value of $\overline{h_2(r)}$ is not needed here, but the expressions for $h_1(r)$ and $h_2(r)$ are given in an appendix.

The rate of transfer of concentration across a plane moving with the mean velocity is Q where

$$\begin{aligned} Q &= a^2\int_0^{2\pi}d\theta\int_0^1 (W_0 w - W)Cr\,dr\\ &= \pi a^2\left(\frac{1}{a}\frac{\partial C_m}{\partial \xi}\right)\frac{a^2W_0^2}{48\kappa}[Q_0+NQ_1+N^2Q_2].\end{aligned}$$

From the above expressions it is found that

$$Q_0 = -1;\quad Q_1 = 0;\quad Q_2 = -\left(\frac{1}{1440}\right)^2\left(-\frac{2569}{33}\sigma^2+\frac{109}{90}\right). \tag{3.15}$$

Since Q is proportional to $\partial C_m/\partial \xi$, the longitudinal diffusivity exists, and is

$$D = -\frac{Q}{\pi a^2\left(\dfrac{1}{a}\dfrac{\partial C_m}{\partial \xi}\right)}$$

$$= \frac{a^2W_0^2}{48\kappa}\left[1+\left(\frac{N}{1440}\right)^2\left(-\frac{2569}{33}\sigma^2+\frac{109}{90}\right)\right]. \tag{3.16}$$

It is probably more convenient to write this in terms of W, the actual mean velocity given by (3.9). We replace W_0 by $W(1+AN^2)$ throughout (except that in N we may simply replace W_0 by W to the order of this work) and obtain

$$D = \frac{a^2W^2}{48\kappa}\left[1+\left(\frac{N}{1440}\right)^2\left(-\frac{2569}{33}\sigma^2+\frac{1797}{70}\right)\right], \tag{3.17}$$

where now $$N = \left(\frac{a}{b}\right)\left(\frac{Wa}{\nu}\right)^2.$$

Some comments on the value of the longitudinal diffusivity

It can be shown that D is an even function of N, so that the next term in (3.17) is of order N^4. The equations obtained from (3.2) by letting $\epsilon \to 0$, and equation (3.11) are invariant, as are the boundary conditions, under the following transformations:

$$N \to -N;\ \theta \to -\theta;\ v \to -v;\ u \to u;\ w \to w;\ p \to p;\ C \to C;\ \xi \to \xi. \tag{3.18}$$

Thus the flux Q with a change in sign of N is

$$Q(-N) = a^2\int_0^{2\pi}\int_0^1 \{W_0 w(r, -\theta; N) - W(-N)\}C(r, -\theta; N)r\,dr\,d\theta$$

$$= a^2\int_0^{2\pi}\int_0^1 \{W_0 w(r, \psi; N) - W(-N)\}C(r, \psi; N)r\,dr\,d\psi$$

$$= Q(N), \tag{3.19}$$

since clearly $W(-N) = W(N)$ from (3.18). Equation (3.19) is still true even when ϵ is not zero, for it can easily be seen that the full equations are invariant under the transformations (3.18), if in addition $\epsilon \to -\epsilon$ and $\tau \to \tau$. The transformations (3.18) have a clear physical interpretation. A change of sign in N can only be interpreted as a change of sign in the curvature of the coil in which the pipe is wound, so that, in figure 2, O is replaced by its mirror image in the plane $\theta = 0$. Clearly the solutions are then obtained by reflexion in the same plane, and this consideration yields (3.18).

The term independent of N in (3.16) and (3.17) is the solution obtained by Taylor, and the additional term gives the effect of the curvature. It can be seen from (3.17) that D is reduced by the curvature only if $\sigma > 0{\cdot}57$.

The explanation of the peculiar effect of a small value of σ is that the change in D caused by the secondary flow is small compared with that due to the change in the axial velocity profile, as can be seen from the equations for C_1 and C_2 in (3.13). If, in the equation for C_1, we compare the orders of the two terms on the right-hand side, using the values previously found for C_0 and Υ_{01} we find that their ratio is of order σ. Clearly the term involving Υ_{01} is the effect of the secondary flow in the cross-sectional plane, and the first term represents the effect of the change in the axial velocity. A similar analysis applies to the equation for C_2. Thus, for small σ, the changes in C of order N, and of order N^2, and so also in D are caused by the change in the axial velocity profile. It is equivalent to argue that, if κ is very large, the lateral mixing is dominated by molecular diffusion, and not by advection with the secondary flow. However, for all common liquids and for most gases, D is reduced by the curvature for the reasons given in the introduction.

The difference between (3.16) and (3.17) is that (3.16) gives the change in D due to pipe curvature when the pressure gradient is kept fixed, and (3.17) that when the mean velocity is kept fixed. The difference is important only if σ is very small.

Although Dean suggests that his expressions for the velocities are valid for N up to about 50, a more severe restriction may arise from the need for D to be positive. This implies that the expression (3.17) cannot be valid unless

$$\left(\frac{1440}{N}\right)^2 > \frac{2569}{33}\sigma^2 - \frac{1797}{70}. \tag{3.20}$$

For a case in which $\sigma = 10^3$ this requires $N < 0{\cdot}163$, so that when $\epsilon = 10^{-2}$, Wa/ν must be less than 4·04. When $\sigma = 1$ the requirement is that $N < 199$, so that when $\epsilon = 10^{-2}$, Wa/ν must be less than 141. These considerations suggest that the formulae are likely to be more useful for diffusion in gases for which σ is generally near 1, than in liquids, for which values of σ are typically of order 10^3. There is the further point that the molecular diffusivity has been assumed to be independent of concentration. This is known to be a much better approximation for gases than for liquids. (This last point applies to all work done on this subject and not just to the present paper.) These remarks are only meant as indications; what is really needed is a radius of convergence of the series of which (3.17) is the first two terms.

4. The effect of buoyancy forces on laminar dispersion in a horizontal pipe

Preliminary remarks

For the reason given in the introduction, the case of flow of a solute in a straight pipe, when the density of the solute is different from that of the ambient fluid, is not one where we expect the longitudinal dispersion of the solute to be governed by a diffusion equation when $t \gg a^2/\kappa$ (although there is the possibility, discussed later, that for very large times, perhaps much greater than those necessary for Taylor's analysis to be valid, the additional dynamical effects due to the buoyancy become negligible and then the Taylor solution may govern the final form of the mean concentration). However, if the local length scale of the distribution of concentration in the axial direction (e.g. $(\partial C/\partial x)/(\partial^2 C/\partial x^2)$) is large compared with Wa^2/κ it is still permissible to calculate the flux of concentration Q across planes moving with the mean velocity assuming that the flow properties are locally uniform and steady (with respect to axes moving with the mean velocity); this is exactly the assumption made by Taylor (1953) and explained in §2. The fact that there is now no longitudinal diffusivity shows itself in the form of Q; it will be found that Q is no longer a linear function of $\partial C_m/\partial X$ (where $X = x - Wt$ and the axes are those of figure 1), but contains terms proportional to all odd powers of $\partial C_m/\partial X$. The equation for the mean concentration that takes the place of the diffusion equation is then found from the continuity equation for the concentration in the same manner and under the same assumptions as in Taylor (1953).

In this work the change in density caused by the concentration will be neglected except in the body force term, that is, except when the change in density is multiplied by g. This approximation is the analogy of the well-known Boussinesq approximation in the case when the density changes are caused by temperature. It will be assumed that the density of the fluid is given by

$$\rho = \rho_0(1+\alpha C),$$

where ρ_0 and α are constants, independent of concentration.

The equations then admit solutions which are exactly steady with respect to axes moving with the mean velocity, and in which $\partial C/\partial X$ and the velocity components are independent of X. If a non-dimensional number G (defined in (4.8) and proportional to $\partial C/\partial X$) is small enough, the dependent variables can be expanded in power series in G. The introduction of the uniform concentration gradient causes the relationship between W, the mean velocity, and

$$\left\{-\frac{1}{\rho_0}\frac{\partial p'}{\partial X}\right\}_{r=0}$$

to be different from that when $G = 0$. For convenience we write

$$\Gamma = \left\{-\frac{1}{\rho_0}\frac{\partial p'}{\partial X}\right\}_{r=0} \Big/ \frac{\nu W}{a^2}, \tag{4.1}$$

and the relationship is, to order G^2,

$$\Gamma = 8(1+\Delta G^2), \tag{4.2}$$

where Δ is a non-dimensional number. A similar relationship was found by Morton (1959) when the change was caused by a uniform temperature gradient.

In our calculations it is convenient for reasons to be discussed later (and there is no loss of generality) to regard the value of W as given, so that the axial pressure gradient depends on G.

The calculation of the concentration flux

We use non-dimensional velocity components (u, v, w) corresponding to the co-ordinates (ra, θ, x) of figure 1 (with scales chosen so that the actual velocity is $(\nu u/a,\ \nu v/a,\ Ww)$). We leave the pressure in its dimensional form p'. With the above assumptions of steadiness with respect to axes moving with the mean velocity and that $\partial C/\partial X$ and the velocity components are independent of X, the equations to be solved are:

$$\left.\begin{aligned}
u\frac{\partial u}{\partial r}+\frac{v}{r}\frac{\partial u}{\partial \theta}-\frac{v^2}{r} &= -\frac{a^2}{\rho_0\nu^2}\frac{\partial p'}{\partial r}+\left(\nabla^2 u-\frac{u}{r^2}-\frac{2}{r^2}\frac{\partial v}{\partial \theta}\right)-\frac{ga^3\cos\theta}{\nu^2}(1+\alpha C),\\
u\frac{\partial v}{\partial r}+\frac{v}{r}\frac{\partial v}{\partial \theta}+\frac{uv}{r} &= -\frac{a^2}{\rho_0\nu^2}\frac{1}{r}\frac{\partial p'}{\partial \theta}+\left(\nabla^2 v-\frac{v}{r^2}+\frac{2}{r^2}\frac{\partial u}{\partial \theta}\right)+\frac{ga^3\sin\theta}{\nu^2}(1+\alpha C),\\
u\frac{\partial w}{\partial r}+\frac{v}{r}\frac{\partial w}{\partial \theta} &= -\frac{a^2}{\rho_0\nu W}\frac{\partial p'}{\partial X}+\nabla^2 w,\\
\frac{\nu}{\kappa}\left(u\frac{\partial C}{\partial r}+\frac{v}{r}\frac{\partial C}{\partial \theta}\right)+\frac{Wa^2}{\kappa}(w-1)\frac{\partial C}{\partial X} &= \nabla^2 C.
\end{aligned}\right\}\qquad(4.3)$$

On differentiating the first two of these equations with respect to X we find that

$$0 = -\frac{\partial}{\partial X}\left(\frac{1}{\rho_0}\frac{\partial p'}{\partial r}\right)-\alpha ga\cos\theta\frac{\partial C}{\partial X},$$

$$0 = -\frac{\partial}{\partial X}\left(\frac{1}{\rho_0 r}\frac{\partial p'}{\partial \theta}\right)+\alpha ga\sin\theta\frac{\partial C}{\partial X}.$$

Thus, using the definition of Γ (equation (4.1)), we find

$$-\frac{1}{\rho_0}\frac{\partial p'}{\partial X} = \frac{\nu W}{a^2}\Gamma+\alpha gar\cos\theta\frac{\partial C}{\partial X},\qquad(4.4)$$

since $\partial C/\partial X$ is constant. As before, the continuity equation is satisfied identically by using a stream function $\Psi(r,\theta)$ so that

$$u = \frac{1}{r}\frac{\partial \Psi}{\partial \theta},\quad v = -\frac{\partial \Psi}{\partial r}.\qquad(4.5)$$

We write
$$C = X\frac{\partial C_m}{\partial X}+a\frac{\partial C_m}{\partial X}F(r,\theta),\qquad(4.6)$$

since, as in §§2 and 3, $\partial C_m/\partial X = \partial C/\partial X$. On eliminating p' and using (4.4), (4.5) and (4.6) we find

$$\left.\begin{aligned}
\nabla^4\Psi+\frac{1}{r}\frac{\partial(\Psi,\nabla^2\Psi)}{\partial(r,\theta)} &= G\left(\frac{\partial F}{\partial r}\sin\theta+\frac{\cos\theta}{r}\frac{\partial F}{\partial \theta}\right),\\
\nabla^2 w+\Gamma+\frac{1}{r}\frac{\partial(\Psi,w)}{\partial(r,\theta)} &= -G\frac{\sigma}{P}r\cos\theta,\\
\nabla^2 F &= P(w-1)-\frac{\sigma}{r}\frac{\partial(\Psi,F)}{\partial(r,\theta)},
\end{aligned}\right\}\qquad(4.7)$$

where
$$G = \frac{\alpha ga^4}{\nu^2}\frac{\partial C_m}{\partial X};\quad P = \frac{Wa}{\kappa};\quad \sigma = \frac{\nu}{\kappa}.\qquad(4.8)$$

The boundary conditions are

$$\left.\begin{array}{ll}
\text{(i)} & \text{as } G\to 0,\ w\to 2(1-r^2),\\
\text{(ii)} & \dfrac{1}{\pi}\displaystyle\int_0^{2\pi}\int_0^1 wr\,dr\,d\theta = 1,\\
\text{(iii)} & w,\ \Psi,\ \partial\Psi/\partial r \text{ and } \partial F/\partial r \text{ are zero at } r = 1,\\
\text{(iv)} & \text{all quantities are finite at } r = 0.
\end{array}\right\}\qquad(4.9)$$

It has not been found possible to obtain an exact solution of the equations in closed form. We shall assume that Ψ, w, F and Γ can all be expanded as power series in G, like

$$\Psi = \Psi_0 + G\Psi_1 + G^2\Psi_2 + \dots \tag{4.10}$$

and evaluate the terms up to the second order. If $G = 0$ the solution is that given in §2, viz.

$$\Psi_0 = 0; \quad w_0 = 2(1-r^2); \quad F_0 = \tfrac{1}{8}P(2r^2-r^4); \quad \Gamma_0 = 8.$$

On substituting the above expressions into the equations, and comparing the coefficients of G and G^2, the following equations are obtained for the first and second order terms:

$$\left.\begin{aligned}
\nabla^2 w_1 + \Gamma_1 &= \frac{1}{r}\frac{\partial\Psi_1}{\partial\theta}\frac{\partial w_0}{\partial r} - \frac{\sigma}{P} r\cos\theta,\\
\nabla^2 w_2 + \Gamma_2 &= \frac{1}{r}\frac{\partial\Psi_2}{\partial\theta}\frac{\partial w_0}{\partial r} - \frac{1}{r}\frac{\partial(\Psi_1, w_1)}{\partial(r,\theta)},\\
\nabla^2 F_1 &= P w_1 + \frac{\sigma}{r}\frac{\partial\Psi_1}{\partial\theta}\frac{\partial F_0}{\partial r},\\
\nabla^2 F_2 &= P w_2 - \frac{\sigma}{r}\frac{\partial(\Psi_1, F_1)}{\partial(r,\theta)} + \frac{\sigma}{r}\frac{\partial\Psi_2}{\partial\theta}\frac{\partial F_0}{\partial r},\\
\nabla^4\Psi_1 &= \frac{\partial F_0}{\partial r}\sin\theta,\\
\nabla^4\Psi_2 + \frac{1}{r}\frac{\partial(\Psi_1, \nabla^2\Psi_1)}{\partial(r,\theta)} &= \frac{\partial F_1}{\partial r}\sin\theta + \frac{1}{r}\frac{\partial F_1}{\partial\theta}\cos\theta.
\end{aligned}\right\} \tag{4.11}$$

The solutions are found to be of the following forms:

$$\left.\begin{aligned}
w_1 &= i_1(r)\cos\theta, & w_2 &= i_2(r) + \overline{i_2(r)}\cos 2\theta,\\
\Psi_1 &= j_1(r)\sin\theta, & \Psi_2 &= j_2(r)\sin 2\theta,\\
F_1 &= k_1(r)\cos\theta, & F_2 &= k_2(r) + \overline{k_2(r)}\cos 2\theta.
\end{aligned}\right\} \tag{4.12}$$

For our purposes the values of $\overline{i_2(r)}, j_2(r)$ and $\overline{k_2(r)}$ are not needed. The values of the other quantities are given in the appendix, together with the value of Γ_2, found from the condition that

$$\frac{1}{\pi}\int_0^{2\pi}\int_0^1 w_2\, r\,dr\,d\theta = 0;$$

in fact $\Gamma_2 = 8\Delta$. The value of Γ_1 is identically zero, establishing the result stated in (4.2).

The flux Q is given by

$$\begin{aligned}
Q &= a^2\int_0^{2\pi}\int_0^1 W(w-1)C\,r\,dr\,d\theta\\
&= Wa^3\frac{\partial C_m}{\partial X}\int_0^{2\pi}\int_0^1 [(w_0-1) + Gw_1 + G^2w_2][F_0 + GF_1 + G^2F_2]\,r\,dr\,d\theta\\
&= \pi a^2\left(\frac{a^2W^2}{48\kappa}\right)\frac{\partial C_m}{\partial X}[Q_0 + GQ_1 + G^2Q_2],
\end{aligned} \tag{4.13}$$

say, to order G^2. It is found from the above expressions that

$$Q_0 = -1; \quad Q_1 = 0; \quad Q_2 = -\left(\frac{1}{2880}\right)^2$$
$$\times\left[-\frac{2569}{8448}P^2\sigma^2 + \frac{19797}{197120}P^2 + \frac{10425}{56}\sigma + 60480\frac{\sigma^2}{P^2}\right]. \tag{4.14}$$

We now wish to apply the above analysis to the case when $\partial C/\partial X$ is not uniform but is slowly varying. Consider an experiment in which a finite quantity of solute is injected at some point in the tube and then spreads out as it is advected downstream by the flow. Far upstream and downstream where G is zero Γ has the value it had before the solute was introduced. Elsewhere, (4.2) shows that Γ is different at different values of X, since G varies with X. Thus W and $\{-(1/\rho_0)(\partial p'/\partial X)\}_{r=0}$ cannot both be independent of X. However, it is easily seen from the equation of continuity ($\nabla . \mathbf{u} = 0$ with the Boussinesq type approximation)† that the mean axial velocity must be independent of X, so that W is equal to its undisturbed value everywhere in the tube; thus the axial pressure gradient and all other flow quantities vary since G varies with X. This is the reason why we chose the mean velocity as given at the beginning of the calculation.

The equation for the conservation of concentration is:

$$\pi a^2 \frac{\partial C_m}{\partial t} = -\frac{\partial Q}{\partial X}. \tag{4.15}$$

We shall assume that (4.14) is approximately true even if $\partial C_m/\partial X$ is not exactly uniform. This is the same assumption as was made by Taylor (1953) for the case $\alpha = 0$, and proved correct by Aris (1956) (see §2). On substitution (4.15) becomes

$$\frac{\partial C_m}{\partial t} = \frac{a^2W^2}{48\kappa}\frac{\partial}{\partial X}\left[\frac{\partial C_m}{\partial X} - \left\{\frac{\alpha g a^4}{\nu^2}\right\}^2 Q_2 \left(\frac{\partial C_m}{\partial X}\right)^3\right], \tag{4.16}$$

where Q_2 is given in (4.14). Equation (4.16) is the equation that replaces the diffusion equation.

Some comments on the form of Q, and equation (4.16)

It has been stated above that Q is an odd function of $\partial C_m/\partial X$ or equivalently that the quantity

$$\frac{Q}{Wa^3(\partial C_m/\partial X)} = S = \int_0^{2\pi}\int_0^1 (w-1)Fr\,dr\,d\theta \tag{4.17}$$

is an even function of G. The proof of this statement is essentially the same as that given in §3, and the details will not be given here. The equations (4.7) and the boundary conditions (4.9) are now invariant under the transformations

$$G \to -G;\ \theta \to \pi - \theta;\ r \to r;\ \Psi \to -\Psi;\ w \to w;\ F \to F;\ \Gamma \to \Gamma. \tag{4.18}$$

---

† If the Boussinesq approximation is not made it is the mass flux and not the mean velocity that is independent of X; this is clearly equivalent and the above argument is unaffected if W is replaced by the constant value of the mass flux. For a calculation in which the Boussinesq approximation is not made see del Casal & Gill (1962).

The physical interpretation of (4.18) is most easily seen if we think of a change of sign in G as being caused by a change of sign in g. It is then clear that the new solution is found by reflexion in the plane $|\theta| = \frac{1}{2}\pi$, as expressed by (4.18). It is physically plausible that a change of sign in g will not change Q, and this is the same as S being an even function of G. It is equivalent to argue that a change of sign in $\partial C_m/\partial X$ will simply change the sign of Q. (These statements are true in our problem but in the similar problem of Couette flow between two parallel infinite planes, with two-dimensional behaviour assumed, Q is not simply an odd or even function of $\partial C_m/\partial X$; this is because the boundary conditions are not homogeneous and there is no symmetry analogous to that of the present problem.)

The range of validity of the present analysis is difficult to estimate; it must presumably involve G being sufficiently small. Since we expect any concentration distribution to be continually spread out as time increases, G should eventually tend to zero everywhere so that there will be a time when the dominant correction to Q is of order G^2. After this time the distribution of mean concentration should be described by (4.16). Since (4.16) is non-linear, it is possible that there are regions in which its solutions have very sharp changes, and that in these regions neither (4.16) nor the analysis leading to it is valid. But if the solutions of (4.16) are such that asymptotically $\partial C_m/\partial X$ tends to zero everywhere then eventually the term involving $(\partial C_m/\partial X)^3$ will be much smaller than that involving $\partial C_m/\partial X$ and then Taylor's solution will become valid. It is clear, as mentioned above, that the time after which the diffusion equation becomes valid may be much greater than that for which Taylor's original analysis is valid, and clearly cannot be smaller.

If the inertia terms are neglected in the first three equations of (4.3), and the calculations repeated it is found that the value of Q_0 is unchanged and the new value of Q_2 is

$$Q_2' = -\left(\frac{1}{2880}\right)^2\left[-\frac{2569}{8448}P^2\sigma^2 + 60480\,\frac{\sigma^2}{P^2}\right], \tag{4.19}$$

that is, it is obtained from Q_2 by omitting the terms of zero and first order in σ. With the aid of (4.19) the significance of the terms in expression (4.14) can be seen. The term in σ^2/P^2 is that found by Taylor (1953; unpublished) and referred to in the introduction. It arises from the change in the axial velocity caused by the term in (4.4) proportional to $\partial C/\partial X$ and acts to increase the dispersion. The term in $P^2\sigma^2$ is negative and represents the lateral mixing effect that arises from the parts of u and v coming from the interaction between viscous forces and buoyancy forces. The extra terms in (4.14) are not important except for σ less than about 10, and represent the effects of the small changes in velocity and concentration caused by the non-linear terms in the Navier–Stokes equations.

For small P, $-Q_2$ is positive and so the dispersion should be greater than that predicted by the Taylor–Aris solution, but as P increases the effect of the secondary flow becomes more important. At a certain critical P, say P_c, the two effects are in balance and as P increases further the dispersion becomes less than that predicted by Taylor. This dependence on P is to be expected from a consideration of the way in which the value of Q will vary when only W is varied. When W is zero, $Q \neq 0$ because of the axial motions set up by the longitudinal concentration gradient. In fact it is clear that for all W this mechanism gives a contribution to Q independent of W (in our calculations this contribution is that arising from the term in (4.14) proportional to σ^2/P^2, whose contribution to Q is easily seen to be independent of W, using (4.13)). However the secondary flow effect does depend on W, for steady lateral variations of concentration (which are responsible for the secondary flow) can persist against the smoothing action of molecular diffusion only if the axial concentration gradient is being advected in the longitudinal direction. This advection (and so lateral variations of concentration and lateral velocities) is dominated by the basic flow if G is sufficiently small, and so increases with W. Thus as W (and so P, since only W is being varied) is increased the lateral mixing will eventually make a contribution to Q greater than that due to the change in the axial velocity. Thus eventually the dispersion should be reduced by the buoyancy. Our calculation and the above arguments confirm the qualitative explanation of Reejhsinghani *et al.* mentioned in §1.

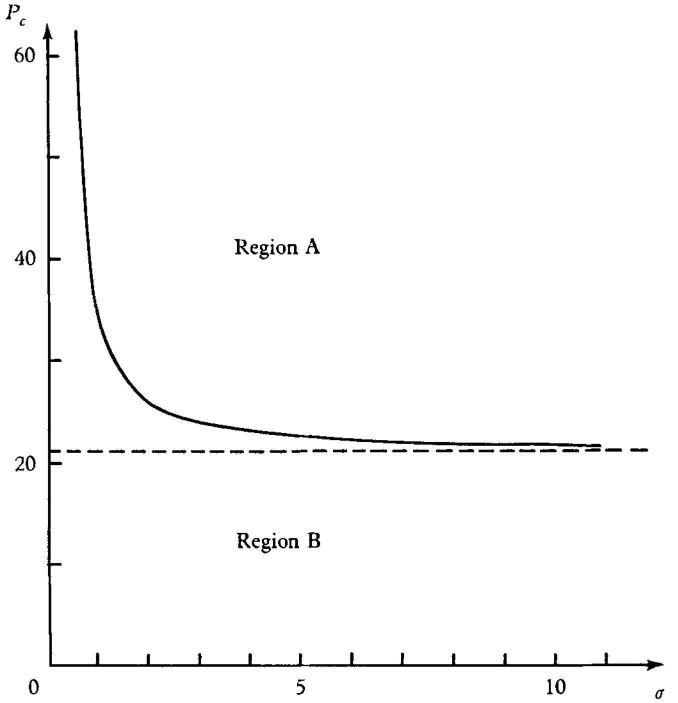

FIGURE 3. The variation of P_c with σ. The dispersion is less than or greater than that predicted by the Taylor-Aris theory according as the point (σ, P) is in region A or region B respectively.

From (4.14) the value of P_c depends only on σ, if G is small enough for our analysis to be valid, and is given by solving the equation $Q_2 = 0$. A graph of P_c against σ is shown in figure 3. For large σ, P_c approaches the constant value given by solving the equation $Q_2' = 0$, that is

$$P_c \rightarrow 21{\cdot}12 \quad \text{as} \quad \sigma \rightarrow \infty. \tag{4.20}$$

It is interesting to compare these results with the observations made by Reejhsinghani *et al.* Their value of σ was 2900 which is large enough for (4.20) to be a very good approximation. In their experiments with the 1·5 mm tube, they observed that for $P = 6{\cdot}25$ the dispersion was 25 % greater than that given by the Taylor–Aris theory, but that for $P = 22{\cdot}5$, which is close to the value of P_c predicted by (4.20), the dispersion was given accurately by the Taylor–Aris theory. Both observations were made at values of $\tau = \kappa t/a^2 \gg 1$. In the experiments with the 5 mm tube, the value of P at which the effect of buoyancy on the dispersion was not noticeable was higher, being between 75 and 125; however the values of τ used in these experiments were much lower, the highest being 2·80 and all the others being less than 1, and since Ananthakrishnan *et al* (1965) showed that the Taylor solution is valid only for $\tau > 1$, it is clear that our analysis is not valid for the values of τ used in the 5 mm tube observations. (But it should be pointed out that Reejhsinghani *et al.* did not compare their experiments with Taylor's solution for $\tau < 1$, but with the numerical solution of Ananthakrishnan *et al.*)

Finally a few remarks on the effect of axial molecular diffusion will be made. In our calculation $\kappa(\partial^2 C/\partial X^2)$ was identically zero, but an equation analogous to (2.9) holds in the general non-steady case provided only that the Boussinesq type approximation is valid. This equation is

$$\frac{\partial C_m}{\partial t} = \kappa \frac{\partial^2 C_m}{\partial X^2} - \frac{\partial}{\partial X}\{W(w-1)C\}_m, \tag{4.21}$$

and is derived by integration of the full concentration equation over the cross-section, using the continuity equation ($\nabla.\mathbf{u} = 0$) and the facts that u, v and $\partial C/\partial r$ are zero at $r = 1$. In §§ 2 and 3 it was possible to use the general analysis of § 1 to show that $\{W(w-1)C\}_m$, which is equal to Q, is asymptotically proportional to $\partial C_m/\partial X$. As explained above this analysis does not now apply. Nevertheless, if the approximation used by Taylor and used here in deriving (4.16) is valid, $\{W(w-1)C\}_m$ is given asymptotically by an expansion of the form of (4.13), with Q_0, Q_1 and Q_2 given in (4.14). Then (4.21) shows that the effect of axial molecular diffusion is independent and additive, as in §§ 2 and 3; thus the right-hand side of (4.16) is modified by an extra term $+\kappa(\partial^2 C_m/\partial X^2)$.

The authors wish to express their gratitude to Prof. G. K. Batchelor for his constant guidance and encouragement during the course of this work, and to Sir Geoffrey Taylor for allowing them to see some of his unpublished work, and for a most helpful discussion. They are also grateful to Prof. W. N. Gill for a copy, prior to publication, of his paper with Reejhsinghani and Barduhn. One of them (M. E. E.) is indebted to the Turkish N.A.T.O. Science Committee and the other (P.C.C.) to the Science Research Council for financial support during the period in which this work was done.

Appendix

Here the values of the functions referred to in §§ 3 and 4 are given:

$$f_1(r) = \frac{1}{72}[-4r + 9r^3 - 6r^5 + r^7],$$

$$g_1(r) = \frac{1}{1440}[19r-40r^3+30r^5-10r^7+r^9],$$

$$g_2(r) = \frac{1}{350\times 576^2}[-4119+21280r^2-46340r^4+55440r^6-39830r^8$$
$$+17584r^{10}-4620r^{12}+640r^{14}-35r^{16}],$$

$$h_1(r) = \frac{1}{300\times 576}[-256r+285r^3-200r^5+75r^7-15r^9+r^{11}]$$
$$+\frac{\sigma}{60\times 576}[-68r+120r^3-130r^5+75r^7-21r^9+2r^{11}],$$

$$h_2(r) = \frac{1}{10368}\left[-\left(\frac{17}{60}\sigma^2+\frac{16}{75}\sigma+\frac{4119}{44800}\right)r^2+\left(\frac{91}{160}\sigma^2+\frac{287}{800}\sigma+\frac{19}{160}\right)r^4\right.$$
$$-\left(\frac{251}{360}\sigma^2+\frac{4901}{14400}\sigma+\frac{331}{2880}\right)r^6+\left(\frac{1129}{1920}\sigma^2+\frac{2033}{9600}\sigma+\frac{99}{1280}\right)r^8$$
$$-\left(\frac{553}{1600}\sigma^2+\frac{37}{400}\sigma+\frac{569}{16000}\right)r^{10}+\left(\frac{259}{1920}\sigma^2+\frac{263}{9600}\sigma+\frac{157}{14400}\right)r^{12}$$
$$-\left(\frac{73}{2240}\sigma^2+\frac{29}{5600}\sigma+\frac{33}{15680}\right)r^{14}+\left(\frac{11}{2560}\sigma^2+\frac{7}{12800}\sigma+\frac{1}{4480}\right)r^{16}$$
$$\left.-\left(\frac{1}{4320}\sigma^2+\frac{1}{43200}\sigma+\frac{1}{103680}\right)r^{18}\right]+\tfrac{1}{4}Ar^2,$$

$$i_1(r) = \frac{P}{32}g_1(r)+\frac{\sigma}{8P}(r-r^3),$$

$$i_2(r) = \frac{P^2}{1024}g_2(r)+\frac{\Gamma_2}{4}(1-r^2)+\frac{\sigma}{40\times 192^2}$$
$$\times[-19+80r^2-130r^4+100r^6-35r^8+4r^{10}],$$

$$j_1(r) = -\frac{P}{32}f_1(r),$$

$$k_1(r) = \frac{P^2}{32}h_1(r)+\frac{\sigma}{192}(-4r+3r^3-r^5),$$

$$k_2(r) = \frac{P^3}{1024}[h_2(r)-\tfrac{1}{4}Ar^2]+\frac{P\Gamma_2}{64}[4r^2-r^4],$$
$$+\frac{P\sigma}{2\times 576^2}\left[-\left(6\sigma+\frac{171}{80}\right)r^2+\left(9\sigma+\frac{9}{4}\right)r^4-\left(\frac{55}{8}\sigma+\frac{13}{8}\right)r^6\right.$$
$$\left.+\left(\frac{93}{32}\sigma+\frac{45}{64}\right)r^8-\left(\frac{27}{40}\sigma+\frac{63}{400}\right)r^{10}+\left(\frac{1}{16}\sigma+\frac{1}{80}\right)r^{12}\right],$$

$$A = \frac{1}{576^2}\frac{3082}{1575},$$

$$\Gamma_2 = \left(\frac{P}{576}\right)^2\frac{1541}{100800}+\frac{\sigma}{576}\frac{11}{960}.$$

REFERENCES

ANANTHAKRISHNAN, V., GILL, W. N. & BARDUHN, A. J. 1965 *A.I.Ch.E. J.* **11**, 1063.

ARIS, R. 1956 *Proc. Roy. Soc.* A **235**, 67.

ARIS, R. 1959 *Proc. Roy. Soc.* A **252**, 538.

BATCHELOR, G. K. & TOWNSEND, A. A. 1956 'Turbulent diffusion' in *Surveys in Mechanics*. Cambridge University Press.

BATCHELOR, G. K. 1966 *Proc. 2nd Australasian Conference on Hydraulics and Fluid Mechanics*, Auckland, Dec. 1965.

BOURNIA, A., COULL, J. & HOUGHTON, G. 1961 *Proc. Roy. Soc.* A **261**, 227.

CARO, C. G. 1966 *J. Physiol.* **185**, 501.

CUMING, H. G. 1952 *A.R.C.R. & M.* no. 2880.

DEAN, W. R. 1927 *Phil. Mag.* (7), **4**, 208.

DEAN, W. R. 1928 *Phil. Mag.* (7), **5**, 673.

DEL CASAL, E. & GILL, W. N. 1962 *A.I.Ch.E. J.* **8**, 570.

EVANS, E. V. & KENNEY, C. N. 1965 *Proc. Roy. Soc.* A **284**, 540.

LIGHTHILL, M. J. 1966 *J. Inst. Maths. Applics.* **2**, 97.

MORTON, B. R. 1959 *Quart. J. Mech. Appl. Math.* **12**, 410.

REEJHSINGHANI, N. S., GILL, W. N. & BARDUHN, A. J. 1966 *A.I.Ch.E. J.* **12**, 916.

TAYLOR, G. I. 1953 *Proc. Roy. Soc.* A **219**, 186.

TAYLOR, G. I. 1954*a* *Proc. Roy. Soc.* A **223**, 446.

TAYLOR, G. I. 1954*b* *Proc. Roy. Soc.* A **225**, 473.

TAYLOR, G. I. 1966 Private communication (but see TAYLOR, G. I. 1929 *Proc. Roy. Soc.* A **124**, 243).

TRUESDELL, L. C. 1963 Ph.D. Thesis. Case Institute of Technology, Cleveland, Ohio, U.S.A.

VAN DEEMTER, J. J., BROEDER, J. J. & LAUWERIER, H. A. 1956 *Appl. Sci. Res.* A **5**, 374.

WHITE, C. M. 1929 *Proc. Roy. Soc.* A **123**, 645.