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The dispersion of a buoyant solute in laminar flow in a straight horizontal pipe. Part 2. The approach to the asymptotic state

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Chatwin (1970) has described the approach to normality of a cloud of solute which is injected into a pipe containing solvent in steady laminar flow. This paper is concerned with the modification of Chatwin's theory when there is a small density difference between the solvent and the dissolved solute. Asymptotic series are derived for the induced density currents and for the distribution of solute in the case when the molecular diffusivity is constant throughout the pipe's cross-section. These series lead to the asymptotic forms of the moments of the distribution, thereby describing some additional deviations from normality caused by buoyancy effects at large times. The theory predicts that the additional dispersion due to buoyancy effects is proportional to the square of the Rayleigh number and depends on the Péclet number of the flow. There is excellent agreement between the results and those previously obtained by the author (1976) from Erdogan & Chatwin's (1967) model of dispersing buoyant solutes. The results confirm that Erdogan & Chatwin's intuitive theory correctly models the significant features of the situation for large Schmidt numbers.

1. Introduction

This paper is concerned with some further aspects of the dispersion of a buoyant solute in a straight horizontal pipe of circular cross-section. In part 1, the author (1976) has presented some calculations based on the earlier analysis of the subject by Erdogan & Chatwin (1967) and both these references contain comprehensive introductory remarks.

To describe the analysis of the problem so far, a buoyant contaminant is injected into a liquid in laminar flow in a straight horizontal pipe, and it subsequently spreads out under the action of density currents, molecular diffusion and axial convection. Erdogan & Chatwin (1967) have derived an equation for the mean concentration $\bar{C}$, replacing the diffusion equation for $\bar{C}$ which holds when the contaminant does not cause dynamical effects. In part 1, the author derived a very rough approximation to $\bar{C}$ and calculated the asymptotic form of the second moment of a distribution of buoyant solute, thereby showing that buoyancy forces at large times have only a small effect on the total dispersion

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of solute. The analysis of part 1 is based on Erdogan & Chatwin's equation for $\bar{C}$, which, in turn, is based on certain physically reasonable (but unjustified) assertions. Clearly it is desirable to justify Erdogan & Chatwin's results.

The two references cited above give a preliminary understanding of the dispersion of buoyant solutes in laminar flow but, as Erdogan & Chatwin remarked, there remains a need for a more detailed treatment of the underlying time-dependent problem. This investigation partially fulfils this need by describing the time-dependent dispersion of a non-passive marker as the asymptotic state is approached.

The technique used in the following work draws on an investigation by Chatwin (1970) of the dispersion of a passive marker at large times. Chatwin showed that a series representation of the form

$$C = T^{-1}C^{(1)}(r,\theta,X) + T^{-2}C^{(2)}(r,\theta,X) + \ldots + T^{-n}C^{(n)}(r,\theta,X) + O(T^{-n-1}) \quad (1.1)$$

is consistent with the concentration equation as $t \to \infty$, where the non-dimensional asymptotic co-ordinates X and T are defined by

$$X = z(\kappa/MW^2a^2t)^{\frac{1}{2}}, \quad T = (Mt\kappa/a^2)^{\frac{1}{2}}. \quad (1.2)$$

The notation in these equations has been introduced in part 1; that is, z is an axial co-ordinate in a frame moving at the discharge speed W and a is the radius of the pipe. At this stage, M may be regarded as a dimensionless constant, whose value is determined later to express the results in the most convenient form. For this part, as in part 1, the molecular diffusivity κ of the solute is assumed to be constant and independent of concentration. Chatwin established that, for a passive marker, the first term in (1.1) was the Gaussian profile originally predicted by Taylor (1953), and that subsequent terms in (1.1) described the deviations from normality at large times. Moreover, Chatwin showed that the series (1.1) was equivalent to several other representations previously obtained for distributions of passive solutes.

The situation is substantially more complicated when the solute gives rise to buoyancy forces. For distributions of buoyant solutes, an asymptotic analysis must include the effects of small axial, radial and azimuthal density currents. This problem is therefore governed by six equations, viz. three Navier–Stokes equations, a concentration equation describing molecular diffusion, and relations describing the fractional rate of expansion and the dependence of density upon concentration. A suitable set of model equations is the set (2.1)–(2.6) in part 1, where, as before, the density depends on concentration according to

$$\rho = \rho_0(1 + \alpha C) \quad (1.3)$$

and the Boussinesq approximation is used. The formula (1.3) should be valid provided that the concentration of the solute is small.

The six governing equations are solved for large times in the following work using asymptotic series to represent the dependent variables C, p, u, v and w. This paper aims to describe how a buoyant cloud of solute injected around $z = z_0$ (say) subsequently disperses under the effects of advection, diffusion and density currents. Now, for exceedingly large times, it is expected that the solute would be dispersed to such an extent that buoyancy forces would be negligible.

This implies that a velocity distribution arbitrarily close to the Poiseuille profile would be obtained by considering t sufficiently large. Thus one would not expect finite density currents as $t \to \infty$ and, moreover, at very large times, the distribution of solute would be determined (as for a passive marker) by a balance between longitudinal advection and cross-sectional molecular diffusion. For this reason, an expansion of the form (1.1) should still be valid for distributions of buoyant solutes. In this proposed expansion, the first term would remain the Gaussian term as for a passive marker, and the subsequent terms would now include the effects of density currents at large times.

The above arguments also suggest that the velocity components need to be represented by the expansions (valid for large t)

$$w \sim w^{(0)} + T^{-1}w^{(1)} + T^{-2}w^{(2)} + \ldots, \tag{1.4}$$

$$u \sim T^{-1}u^{(1)} + T^{-2}u^{(2)} + \ldots, \tag{1.5}$$

$$v \sim T^{-1}v^{(1)} + T^{-2}v^{(2)} + \ldots \ . \tag{1.6}$$

Finally, an expansion for the pressure enables the governing equations to be solved sequentially by equating the coefficients of T^{-n} to zero ($n = 0, 1, 2, \ldots$). The proposed expansion for p is slightly more complicated than those above and is deferred until §2. The determination of the coefficients in these series establishes representations for the dependent variables at large times although no attempt is made to *prove* that the theory is asymptotically correct as $t \to \infty$.

There are two principal conclusions which emerge from this study. The first conclusion is negative in that the dispersion of a buoyant solute cannot be predicted without the full numerical solution of the governing equations. The difficulty precluding a predictive study is that the expansion (1.1) for the concentration profile involves a sequence of arbitrary constants whose values cannot be determined without numerical work. This conclusion reverses that of Chatwin (1970), who found that, for passive markers, a corresponding sequence of constants could be determined analytically by the asymptotic solution of a linear equation.

The second conclusion concerns the accuracy of the results derived in part 1. It is found that there is excellent agreement between the results of parts 1 and 2 provided that the Schmidt number σ of the flow is large.† To summarize the important features of the work, the dispersion induced by buoyancy effects at large times is relatively small, being only $O(t^{-1})$ as $t \to \infty$, whereas the dispersion induced at times when transient effects are significant is $O(t^0)$. The dispersion induced at large times is proportional to the square of the Rayleigh number R, and also depends on the Péclet number P of the flow.† The dispersion is increased in flows for which P is small, and decreased in flows with large values of P. Buoyant and passive markers are found to disperse similarly in flows where P is near a critical Péclet number P_c.

The results of part 2 confirm that the equation for $\bar{C}$ derived by Erdogan & Chatwin (1967) is accurate for large Schmidt numbers, even though it was

† These numbers were defined in equation (2.14) of part 1.

derived from a simplified mathematical model (described in §2 of part 1). This justifies the assumptions incorporated in Erdogan & Chatwin's model.

The plan of the paper is as follows. First some necessary preliminary work is described in §§2 and 3. Series representation of the dependent variables at large times is shown to be consistent with the non-dimensionalized equations. The equations for the coefficients in the series are then solved sequentially to give an approximation to the density currents at large times, and functional forms are established for the coefficients $C^{(n)}$, $n \geqslant 1$, in (1.1). This complicated preparation enables the most important results of the paper to be derived in §4. Here, the terms $C^{(n)}$, $n \geqslant 1$, are calculated explicitly and used to derive the asymptotic form of the moments of the distribution. From the representation of the moments, it is then possible to identify the dispersion which is induced by buoyancy effects at intermediate and at asymptotically large times (§5).

2. Equations for the dependent variables

If the non-dimensional variables w', u', v', r' and p' are defined by

$$w = Ww', \quad u = \nu a^{-1}u', \quad v = \nu a^{-1}v', \quad r = ar', \quad p = \rho_0 W^2 p', \tag{2.1}$$

and the asymptotic co-ordinates X and T are introduced, the system of equations (2.1)–(2.5) in part 1 becomes (now omitting the dashes on the non-dimensionalized variables)

$$\tfrac{1}{2}MT^{-1}\frac{\partial u}{\partial T}-\tfrac{1}{2}MXT^{-2}\frac{\partial u}{\partial X}+T^{-1}w\frac{\partial u}{\partial X}+\sigma\left(u\frac{\partial u}{\partial r}+\frac{v}{r}\frac{\partial u}{\partial \theta}-\frac{v^2}{r}\right)$$
$$= -\frac{P^2}{\sigma}\frac{\partial p}{\partial r}+\frac{\sigma}{P^2}T^{-2}\frac{\partial^2 u}{\partial X^2}+\sigma\left(\nabla^2 u-\frac{u}{r^2}-\frac{2}{r^2}\frac{\partial v}{\partial \theta}\right)-R(1+\alpha C)\cos\theta, \tag{2.2}$$

$$\tfrac{1}{2}MT^{-1}\frac{\partial v}{\partial T}-\tfrac{1}{2}MXT^{-2}\frac{\partial v}{\partial X}+T^{-1}w\frac{\partial v}{\partial X}+\sigma\left(u\frac{\partial v}{\partial r}+\frac{v}{r}\frac{\partial v}{\partial \theta}+\frac{uv}{r}\right)$$
$$= -\frac{P^2}{\sigma r}\frac{\partial p}{\partial \theta}+\frac{\sigma}{P^2}T^{-2}\frac{\partial^2 v}{\partial X^2}+\sigma\left(\nabla^2 v+\frac{2}{r^2}\frac{\partial u}{\partial \theta}-\frac{v}{r^2}\right)+R(1+\alpha C)\sin\theta, \tag{2.3}$$

$$\tfrac{1}{2}MT^{-1}\frac{\partial w}{\partial T}-\tfrac{1}{2}MXT^{-2}\frac{\partial w}{\partial X}+T^{-1}w\frac{\partial w}{\partial X}+\sigma\left(u\frac{\partial w}{\partial r}+\frac{v}{r}\frac{\partial w}{\partial \theta}\right)$$
$$= -T^{-1}\frac{\partial p}{\partial X}+\frac{\sigma}{P^2}T^{-2}\frac{\partial^2 w}{\partial X^2}+\sigma\nabla^2 w, \tag{2.4}$$

$$\tfrac{1}{2}MT^{-1}\frac{\partial C}{\partial T}-\tfrac{1}{2}MXT^{-2}\frac{\partial C}{\partial X}+T^{-1}w\frac{\partial C}{\partial X}+\sigma\left(u\frac{\partial C}{\partial r}+\frac{v}{r}\frac{\partial C}{\partial \theta}\right)$$
$$= \frac{1}{P^2}T^{-2}\frac{\partial^2 C}{\partial X^2}+\nabla^2 C, \tag{2.5}$$

$$T^{-1}\frac{\partial w}{\partial X}+\sigma\left(\frac{1}{r}\frac{\partial}{\partial r}(ru)+\frac{1}{r}\frac{\partial v}{\partial \theta}\right) = 0. \tag{2.6}$$

The parameters P, σ and R were introduced in part 1 [equation (2.14)]. In the above equations, the formula (1.3) has been used to describe the dependence of density upon concentration, and the Boussinesq approximation has been applied.

Now it has been argued in § 1 that the basic flow reverts to Poiseuille flow for exceedingly large times when the solute can be regarded as fully dispersed. In this case, the axial Navier–Stokes equation reduces to the simple form

$$G^* = \frac{\partial p}{\partial z} = \nu\rho\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial w}{\partial r}\right), \tag{2.7}$$

where G^* is a dimensional constant depending on the flow conditions. This equation gives the leading term in an expansion for p, and it follows that a general expansion for p using the asymptotic variables X and T will be

$$p \sim GTX + (\sigma/P)^2\{p^{(0)}(r,\theta,X) + T^{-1}p^{(1)}(r,\theta,X) + \ldots\} \quad \text{for } t \text{ large.} \tag{2.8}$$

Here G is a non-dimensional constant related to G^* by

$$G = G^*a^2/(\rho_0\kappa W).$$

The multiplying factor $(\sigma/P)^2$ is introduced into (2.8) since the appropriate dimensional scaling factor for those pressure forces which cause density currents is

$$\rho_0(\nu/a)^2 = \rho_0 W^2(\nu/Wa)^2 = \rho_0 W^2(\sigma/P)^2,$$

and not $\rho_0 W^2$ as used in (2.1). If it is assumed that ν is constant, the first term in the expansion (1.4) for w is found to be the Poiseuille profile

$$w^{(0)} = 1 - 2r^2 \tag{2.9}$$

when the requirement [see equation (2.27) below]

$$\overline{w^{(0)}} = 0 \tag{2.10}$$

is satisfied.† For (2.10) to hold, G must satisfy the relation

$$G/\sigma = -8. \tag{2.11}$$

The expansion (1.4) for w requires a similar modification to ensure that w is correctly scaled; that is, write

$$w \sim w^{(0)} + (\sigma/P)\{T^{-1}w^{(1)} + T^{-2}w^{(2)} + \ldots\}, \tag{2.12}$$

where the factor σ/P has been introduced because the axial density currents should be scaled by

$$\nu/a = W\sigma/P,$$

and not W as in (2.1).

Equations for the dependent variables are now obtained by substituting the expansions (1.1), (2.8), (1.5), (1.6) and (2.12) into (2.2)–(2.6) and equating the coefficients of T^{-n} to zero. Thus the equations from the T^0 coefficients are

$$0 = -\sigma\,\partial p^{(0)}/\partial r - R\cos\theta, \tag{2.13}$$

$$0 = -\frac{\sigma}{r}\frac{\partial p^{(0)}}{\partial\theta} + R\sin\theta, \tag{2.14}$$

$$0 = -G + \frac{\sigma}{r}\frac{\partial}{\partial r}\left(r\frac{\partial w^{(0)}}{\partial r}\right), \tag{2.15}$$

† An overbar denotes the cross-sectional mean.

and those from the $O(T^{-1})$ terms are

$$0 = -\sigma p_r^{(1)} + \sigma\left(\nabla^2 u^{(1)} - \frac{u^{(1)}}{r^2} - \frac{2}{r^2}\frac{\partial v^{(1)}}{\partial\theta}\right) - R\alpha C^{(1)}\cos\theta, \tag{2.16}$$

$$0 = -\frac{\sigma}{r}p_\theta^{(1)} + \sigma\left(\nabla^2 v^{(1)} + \frac{2}{r^2}\frac{\partial u^{(1)}}{\partial\theta} - \frac{v^{(1)}}{r^2}\right) + R\alpha C^{(1)}\sin\theta, \tag{2.17}$$

$$\sigma u^{(1)} w_r^{(0)} = -\left(\frac{\sigma}{P}\right)^2 p_X^{(0)} + \frac{\sigma^2}{P}\nabla^2 w^{(1)}, \tag{2.18}$$

$$0 = \nabla^2 C^{(1)}, \tag{2.19}$$

$$\sigma\left(\frac{1}{r}\frac{\partial}{\partial r}(ru^{(1)}) + \frac{1}{r}\frac{\partial v^{(1)}}{\partial\theta}\right) = 0. \tag{2.20}$$

The general equations from the coefficients of T^{-n} for $n \geqslant 2$ are

$$\begin{aligned}-\tfrac{1}{2}M(n-2)\,u^{(n-2)} - \tfrac{1}{2}MXu_X^{(n-2)} + w^{(0)}u_X^{(n-1)} + \frac{\sigma}{P}\sum_{j=1}^{n-2} w^{(j)}u_X^{(n-1-j)} \\ + \sigma\sum_{j=1}^{n-1}\left\{u^{(j)}u_r^{(n-j)} + \frac{1}{r}v^{(j)}u_\theta^{(n-j)} - \frac{1}{r}v^{(j)}v^{(n-j)}\right\} \\ = -\sigma p_r^{(n)} + \frac{\sigma}{P^2}u_{XX}^{(n-2)} + \sigma\left(\nabla^2 u^{(n)} - \frac{u^{(n)}}{r^2} - \frac{2}{r^2}\frac{\partial v^{(n)}}{\partial\theta}\right) - R\alpha C^{(n)}\cos\theta,\end{aligned} \tag{2.21}$$

$$\begin{aligned}-\tfrac{1}{2}M(n-2)\,v^{(n-2)} - \tfrac{1}{2}MXv_X^{(n-2)} + w^{(0)}v_X^{(n-1)} + \frac{\sigma}{P}\sum_{j=1}^{n-2} w^{(j)}v_X^{(n-1-j)} \\ + \sigma\sum_{j=1}^{n-1}\left\{u^{(j)}v_r^{(n-j)} + \frac{1}{r}v^{(j)}v_\theta^{(n-j)} + \frac{1}{r}u^{(j)}v^{(n-j)}\right\} \\ = -\frac{\sigma}{P}p_\theta^{(n)} + \frac{\sigma}{P^2}v_{XX}^{(n-2)} + \sigma\left(\nabla^2 v^{(n)} + \frac{2}{r^2}\frac{\partial u^{(n)}}{\partial\theta} - \frac{v^{(n)}}{r^2}\right) + R\alpha C^{(n)}\sin\theta,\end{aligned} \tag{2.22}$$

$$\begin{aligned}-\tfrac{1}{2}M(n-2)\,w^{(n-2)} - \tfrac{1}{2}MXw_X^{(n-2)} + w^{(0)}w_X^{(n-1)} + \frac{\sigma}{P}\sum_{j=1}^{n-2} w^{(j)}w_X^{(n-1-j)} \\ + P\left\{u^{(n)}w_r^{(0)} + \frac{1}{r}v^{(n)}w_\theta^{(0)}\right\} + \sigma\sum_{j=1}^{n-1}\left\{u^{(j)}w_r^{(n-j)} + \frac{1}{r}v^{(j)}w_\theta^{(n-j)}\right\} \\ = -\frac{\sigma}{P}p_X^{(n-1)} + \frac{\sigma}{P^2}w_{XX}^{(n-2)} + \sigma\nabla^2 w^{(n)},\end{aligned} \tag{2.23}$$

$$\begin{aligned}-\tfrac{1}{2}M(n-2)\,C^{(n-2)} - \tfrac{1}{2}MXC_X^{(n-2)} + w^{(0)}C_X^{(n-1)} + \frac{\sigma}{P}\sum_{j=1}^{n-2} w^{(j)}C_X^{(n-1-j)} \\ + \sigma\sum_{j=1}^{n-1}\left\{u^{(j)}C_r^{(n-j)} + \frac{1}{r}v^{(j)}C_\theta^{(n-j)}\right\} = \frac{1}{P^2}\frac{\partial^2 C^{(n-2)}}{\partial X^2} + \nabla^2 C^{(n)},\end{aligned} \tag{2.24}$$

$$\frac{\sigma}{P}w_X^{(n-1)} + \sigma\left(\frac{1}{r}\frac{\partial}{\partial r}(ru^{(n)}) + \frac{1}{r}\frac{\partial v^{(n)}}{\partial\theta}\right) = 0. \tag{2.25}$$

Since the terms in each of the asymptotic series are independent, the above equations are to be solved under the boundary conditions

$$\left.\begin{aligned} W + Ww^{(0)} &= 0 \\ u^{(i)} = v^{(i)} = w^{(i)} &= 0, \quad i = 1, 2, \ldots, \\ \partial C^{(i)}/\partial r &= 0, \quad i = 1, 2, \ldots, \end{aligned}\right\} \quad \text{at} \quad r = 1 \tag{2.26}$$

with the further condition, holding in the frame of reference moving at the discharge speed,

$$\overline{w^{(i)}} = 0, \quad i = 1, 2, \ldots. \tag{2.27}$$

There are two other conditions which must be imposed on the solutions: the velocity distributions must always be finite and the concentration profile must be such that

$$\iiint_{\text{whole pipe}} Z^p C^{(i)} r\, dr\, d\theta\, dz < \text{constant}, \quad i = 1, 2, \ldots, \quad p = 0, 1, 2, \ldots. \tag{2.28}$$

This ensures the existence of all the moments of the distribution.

3. The successive representation of the dependent variables

The equations derived in §2 are now solved successively to establish the asymptotic series for the dependent variables. Only the first two sets of equations are considered in depth in this section (in §3.1) and routine details of further calculations for the density currents are suppressed or included in the appendix. The concentration equation (2.24) is systematically investigated in the last half of the section (§3.2).

3.1. *The velocity components*

The first set of equations (2.13)–(2.15) has the solutions

$$p^{(0)}(r, \theta, X) = (-R/\sigma)\, r \cos\theta + \pi^{(0)}(X), \tag{3.1}$$

$$w^{(0)}(r) = 1 - 2r^2, \tag{3.2}$$

where $\pi^{(0)}$ is a function of X to be determined by applying (2.27) to higher approximations. In the second set of equations (2.16)–(2.20), the continuity equation (2.20) is satisfied identically by using a stream function $\psi^{(1)}(r, \theta, X)$ such that

$$u^{(1)} = r^{-1}\partial\psi^{(1)}/\partial\theta, \quad v^{(1)} = -\partial\psi^{(1)}/\partial r \tag{3.3}$$

and which, using (2.26), satisfies the boundary conditions

$$\psi^{(1)} = \partial\psi^{(1)}/\partial r = 0 \quad \text{at} \quad r = 1. \tag{3.4}$$

With this stream function, $p^{(1)}$ may be eliminated from (2.16) and (2.17), giving

$$\sigma\nabla^4\psi^{(1)} = R\alpha \frac{\partial C^{(1)}}{\partial r} \sin\theta. \tag{3.5}$$

But since it follows from (2.19) and (2.26) that

$$C^{(1)} = f^{(1)}(X), \tag{3.6}$$

(3.4) and (3.5) only admit the trivial solution $\psi^{(1)} = 0$. (The unknown function $f^{(1)}(X)$ in (3.6) is determined later by applying a consistency condition to the equation for $C^{(3)}$ in the manner of Chatwin 1970.) Also the equation for $w^{(1)}$ now takes the form

$$\nabla^2 w^{(1)} = P^{-1}\pi_X^{(0)},$$

with the solution

$$w^{(1)}(r,\theta,X) = P^{-1}\pi_X^{(0)}\tfrac{1}{4}(r^2-1).$$

The application of (2.27) thus gives a first-order equation for $\pi^{(0)}$ which implies that the first-order axial velocity perturbation vanishes; i.e.

$$w^{(1)}(r,\theta,X) = 0. \tag{3.7}$$

These results show that the buoyant solute only induces $O(T^{-2})$ velocities as $t\to\infty$. The discussion of the $O(T^{-1})$ terms is completed by observing that either (2.16) or (2.17) implies

$$p^{(1)}(r,\theta,X) = (-\alpha R/\sigma)\, C^{(1)} r\cos\theta + \pi^{(1)}(X), \tag{3.8}$$

where $\pi^{(1)}(X)$ is to be determined in the same manner as $\pi^{(0)}(X)$.

The set of equations resulting from $O(T^{-2})$ terms may be solved in a similar way. Since $w^{(1)} = 0$, (2.25) admits a stream function $\psi^{(2)}$ with properties analogous to (3.3) and (3.4) and which satisfies the equation

$$\sigma\nabla^4\psi^{(2)} = \alpha R\frac{\partial C^{(2)}}{\partial r}\sin\theta. \tag{3.9}$$

The equation for $C^{(2)}$ becomes

$$\nabla^2 C^{(2)} = w^{(0)}C_X^{(1)}, \tag{3.10}$$

with a solution of the form

$$C^{(2)}(r,X) = g^{(2)}(r)\, df^{(1)}/dX + f^{(2)}(X). \tag{3.11}$$

Here $f^{(2)}(X)$ is an unknown function of X which is determined later, and $g^{(2)}(r)$ satisfies the equations

$$\left.\begin{aligned} \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial g^{(2)}}{\partial r}\right) &= w^{(0)}, \\ [\partial g^{(2)}/\partial r]_{r=1} = 0, \quad \overline{g^{(2)}} &= 0. \end{aligned}\right\} \tag{3.12}$$

The equations for $g^{(2)}$, $\psi^{(2)}$ and $w^{(2)}$ may be solved successively to give the representations

$$g^{(2)}(r) = \tfrac{1}{24}(-3r^4+6r^2-2), \tag{3.13}$$

$$\psi^{(2)}(r,\theta,X) = -\frac{\alpha R}{\sigma}\frac{df^{(1)}}{dX}\sin\theta\,\frac{1}{2304}(r^7-6r^5+9r^3-4r), \tag{3.14}$$

$$\begin{aligned} w^{(2)}(r,\theta,X) = \alpha R\frac{P}{\sigma}\frac{df^{(1)}}{\partial X}\cos\theta\Big[&\frac{1}{8P^2}(-r^3+r) \\ &+\frac{1}{46080}\frac{1}{\sigma}(r^9-10r^7+30r^5-40r^3+19r)\Big]. \end{aligned} \tag{3.15}$$

Higher terms in the series for the velocity variables and the pressure may be found quite readily by systematically solving the equations derived in §2.

$u \sim T^{-2}u^{(2)}+T^{-3}u^{(3)}+\ldots$	$v \sim T^{-2}v^{(2)}+T^{-3}v^{(3)}+\ldots$
$u^{(2)} = k_1(r)\cos\theta$	$v^{(2)} = l_1(r)\sin\theta$
$u^{(3)} = k_2(r)\cos\theta$	$v^{(3)} = l_2(r)\sin\theta$
$u^{(4)} = k_3(r)\cos\theta+k_4(r)\cos 2\theta$	
$u^{(5)} = k_5(r)+k_6(r)\cos\theta+k_7(r)\cos 2\theta$	
$w \sim w^{(0)}(r)+(\sigma/P)\{T^{-2}w^{(2)}+T^{-3}w^{(3)}+\ldots\}$	$p \sim GTX+(\sigma/P)^2\{p^{(0)}+T^{-1}p^{(1)}+\ldots\}$
$w^{(0)} = 1-2r^2$	$p^{(0)} = n_0(r)\cos\theta$
$w^{(2)} = m_1(r)\cos\theta$	$p^{(1)} = n_1(r)\cos\theta$
$w^{(3)} = m_2(r)\cos\theta$	$p^{(2)} = n_2(r)\cos\theta$
$w^{(4)} = m_3(r)+m_4(r)\cos\theta+m_5(r)\cos 2\theta$	$p^{(3)} = \pi^{(3)}(X)+n_3(r)\cos\theta$

TABLE 1. The form of the coefficients in the non-dimensional asymptotic series for u, v, w and p for large t. Some further details are given in the appendix.

There is one slight complication since a stream-function approach is no longer applicable for terms $O(T^{-n})$ when $n \geqslant 3$; the method used to obtain the velocity variables $u^{(n)}$ and $v^{(n)}$ for $n \geqslant 3$ has been described briefly in the appendix. The terms in the series for u, v, w and p have been summarized in table 1 with the important coefficient functions listed in the appendix. The remaining functions mentioned in table 1 are not required since it is found that they do not contribute to the profile $\bar{C}$.

3.2. *The determination of the coefficients for* $\bar{C}$

The coefficient $C^{(2)}$ is given by (3.11) and (3.12) in §3.1. When $n = 3$, (2.24) becomes

$$-\tfrac{1}{2}MC^{(1)}-\tfrac{1}{2}MXC^{(1)}_X+w^{(0)}C^{(2)}_X-P^{-2}C^{(1)}_{XX} = \nabla^2 C^{(3)}, \tag{3.16}$$

whence a differential equation for $C^{(1)}(X)$ (or $f^{(1)}(X)$) is obtained by demanding that both sides have the same cross-sectional mean. It follows using (2.26) that the equation for $f^{(1)}$ is

$$\frac{2}{M}\left\{\frac{1}{P^2}-\overline{w^{(0)}g^{(2)}}\right\}\frac{d^2f^{(1)}}{dX^2}+X\frac{df^{(1)}}{dX}+f^{(1)} = 0, \tag{3.17}$$

the same equation as was established by Chatwin (1970) for the leading term in the profile of a passive marker (and consistent with the remarks made in §1). It is clear immediately that the simplest representation for the distribution will use a co-ordinate X in which M is chosen to be

$$M = 2\{P^{-2}-\overline{w^{(0)}g^{(2)}}\}. \tag{3.18}$$

The previous three equations now give for $C^{(3)}$

$$\nabla^2 C^{(3)} = \{w^{(0)}g^{(2)}-\overline{w^{(0)}g^{(2)}}\}f^{(1)}_{XX}+w^{(0)}f^{(2)}_X,$$

and this equation has the solution

$$C^{(3)} = g^{(3)}(r)f^{(1)}_{XX}+g^{(2)}(r)f^{(2)}_X+f^{(3)}(X). \tag{3.19}$$

Here $f^{(3)}$ is another (as yet) arbitrary function of X, $g^{(2)}$ is determined by (3.12) and $g^{(3)}$ satisfies the equations

$$\left.\begin{aligned}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial g^{(3)}}{\partial r}\right) &= w^{(0)}g^{(2)} - \overline{w^{(0)}g^{(2)}},\\ [\partial g^{(3)}/\partial r]_{r=1} = 0, &\quad \overline{g^{(3)}} = 0.\end{aligned}\right\} \tag{3.20}$$

For $n = 4$, (2.24) gives an equation for $C^{(4)}$:

$$\nabla^2 C^{(4)} = -MC^{(2)} - \tfrac{1}{2}MXC^{(2)}_X + w^{(0)}C^{(3)}_X + \frac{\sigma}{P}\sum_{j=1}^{2} w^{(j)}C^{(3-j)}_X + \sigma\sum_{j=1}^{3}\left\{u^{(j)}C^{(4-j)}_r + \frac{1}{r}v^{(j)}C^{(4-j)}_\theta\right\} - \frac{1}{P^2}C^{(2)}_{XX}, \tag{3.21}$$

for which, after substitution for the terms on the right-hand side, the condition for identical cross-sectional means of both sides leads to the equation

$$f^{(2)}_{XX} + Xf^{(2)}_X + 2f^{(2)} = (2/M)\,\overline{w^{(0)}g^{(3)}}f^{(1)}_{XXX}. \tag{3.22}$$

This enables (3.21) to be written as

$$\nabla^2 C^{(4)} = \left\{\frac{\sigma}{P}w^{(2)} + \sigma u^{(2)}\frac{dg^{(2)}}{dr}\right\}f^{(1)}_X + \{w^{(0)}g^{(3)} - \overline{w^{(0)}g^{(3)}} - g^{(2)}\overline{w^{(0)}g^{(2)}}\}f^{(1)}_{XXX} + \{w^{(0)}g^{(2)} - \overline{w^{(0)}g^{(2)}}\}f^{(2)}_{XX} + w^{(0)}f^{(3)}_X, \tag{3.23}$$

which has a solution of the form

$$C^{(4)}(r,\theta,X) = g^{(4,1)}(r)f^{(1)}_{XXX} + g^{(4,2)}(r,\theta,X)f^{(1)}_X + g^{(3)}(r)f^{(2)}_{XX} + g^{(2)}(r)f^{(3)}_X + f^{(4)}(X), \tag{3.24}$$

where $g^{(2)}$ and $g^{(3)}$ are as defined previously and $g^{(4,1)}$ and $g^{(4,2)}$ are solutions of the problems

$$\left.\begin{aligned}\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial g^{(4,1)}}{\partial r}\right) &= w^{(0)}g^{(3)} - \overline{w^{(0)}g^{(3)}} - g^{(2)}\overline{w^{(0)}g^{(2)}},\\ [\partial g^{(4,1)}/\partial r]_{r=1} = 0, &\quad \overline{g^{(4,1)}} = 0,\end{aligned}\right\} \tag{3.25}$$

$$\left.\begin{aligned}\nabla^2 g^{(4,2)} &= \frac{\sigma}{P}w^{(2)} + \sigma u^{(2)}\frac{dg^{(2)}}{dr},\\ [\partial g^{(4,2)}/\partial r]_{r=1} = 0, &\quad \overline{g^{(4,2)}} = 0.\end{aligned}\right\} \tag{3.26}$$

The term $g^{(4,2)}(r,\theta,X)f^{(1)}_X$ occurs in (3.24) because of the density currents induced by the buoyant solute, and $f^{(4)}$ is a further unknown function of X.

The analysis of (2.24) may be continued to as high a level of approximation as desired, and further results are merely recorded below without including the cumbersome algebraic details. Many of the inhomogeneous terms in the equations for the $f^{(j)}(X)$, $j \geqslant 3$, have been omitted below since they are the zero cross-sectional means of terms of the form func. $(r)\cos n$. It is found that $C^{(5)}$ has the form

$$C^{(5)}(r,\theta,X) = g^{(5,1)}(r)f^{(1)}_{XXXX} + g^{(5,2)}(r,\theta,X)f^{(1)}_{XX} + g^{(5,3)}(r,\theta,X)f^{(1)}_X + g^{(4,1)}(r)f^{(2)}_{XXX} + g^{(4,2)}(r,\theta,X)f^{(2)}_X + g^{(3)}(r)f^{(3)}_{XX} + g^{(2)}(r)f^{(4)}_X + f^{(5)}(X) \tag{3.27}$$

and $f^{(3)}, f^{(4)}$ and $f^{(5)}$ satisfy the equations

$$f^{(3)}_{XX} + Xf^{(3)}_X + 3f^{(3)} = \frac{2}{M}\overline{w^{(0)}g^{(4,1)}}f^{(1)}_{XXXX} + \frac{2}{M}\overline{w^{(0)}g^{(3)}}f^{(2)}_{XXX}, \quad (3.28)$$

$$f^{(4)}_{XX} + Xf^{(4)}_X + 4f^{(4)} = \frac{2}{M}\overline{w^{(0)}g^{(5,1)}}f^{(1)}_{XXXXX} + \frac{2}{M}\overline{w^{(0)}g^{(4,1)}}f^{(2)}_{XXXX} + \frac{2}{M}\overline{w^{(0)}g^{(3)}}f^{(3)}_{XXX}, \quad (3.29)$$

$$\begin{aligned} f^{(5)}_{XX} + Xf^{(5)}_X + 5f^{(5)} = {} & \frac{2}{M}\overline{w^{(0)}g^{(6,1)}}f^{(1)}_{XXXXXX} + \frac{2}{M}\overline{w^{(0)}g^{(5,1)}}f^{(2)}_{XXXXX} \\ & + \frac{2}{M}\overline{w^{(0)}g^{(4,1)}}f^{(3)}_{XXXX} + \frac{2}{M}\overline{w^{(0)}g^{(3)}}f^{(4)}_{XXX} + \frac{2}{M}f^{(1)}_{XX}\left\{\overline{w^{(0)}g^{(6,4)}} + \frac{\sigma}{P}\left(\overline{w^{(2)}g^{(4,2)}} + \overline{w^{(4)}g^{(2)}}\right)\right\} \\ & + \frac{2}{M}f^{(1)}_X\left\{\overline{w^{(0)}\frac{\partial g^{(6,4)}}{\partial X}} + \frac{\sigma}{P}\overline{w^{(2)}\frac{\partial g^{(4,2)}}{\partial X}} + \sigma\left(\overline{u^{(3)}\frac{\partial g^{(4,2)}}{\partial r}} + \overline{\frac{1}{r}v^{(3)}\frac{\partial g^{(4,2)}}{\partial \theta}} + \overline{u^{(5)}\frac{\partial g^{(2)}}{\partial r}}\right)\right\}. \end{aligned} \quad (3.30)$$

The above analysis of (2.24) is sufficient to determine the contributions of large-time density currents to the variance and skewness of a buoyant cloud of solute. The equations for the $f^{(j)}(X)$ are similar to the corresponding equations derived by Chatwin (1970) for a passive marker and only $f^{(5)}$ has additional terms caused by buoyancy effects. Equations defining the coefficient functions $g^{(r,s)}(r, \theta, X)$, $r \geqslant 4$, $s \geqslant 2$, are given in the appendix, which also contains the form of these functions where this is significant.

4. The asymptotic form of the concentration profile

The differential equations derived above for the functions $f^{(j)}(X)$, $j = 1, \ldots, 5$, are solved in this section. This leads to a formal asymptotic representation for the concentration profile which can be used to establish the asymptotic behaviour of the moments of the distribution. The following work uses the Hermite polynomials $\mathrm{He}_n(X)$ in the expressions for the $f^{(j)}(X)$, these polynomials being defined by Rodrigues' formula

$$\mathrm{He}_n(X) = (-1)^n \exp(\tfrac{1}{2}X^2)\, d^n \exp(-\tfrac{1}{2}X^2)/dX^n. \quad (4.1)$$

The equation (3.17) for $f^{(1)}(X)$ [with the constant M defined by (3.18)] has the acceptable solution

$$f^{(1)}(X) = \beta^{(1,0)}\mathrm{He}_0(X)\exp(-\tfrac{1}{2}X^2), \quad (4.2)$$

in which (2.28) has been used in rejecting another independent solution. From (3.22), the general solution for $f^{(2)}(X)$ which does not violate (2.28) is seen to be

$$f^{(2)}(X) = \{\beta^{(2,0)}\mathrm{He}_1(X) + \beta^{(2,1)}\mathrm{He}_3(X)\}\exp(-\tfrac{1}{2}X^2), \quad (4.3)$$

where the second term is a particular solution provided that $\beta^{(2,1)}$ is defined by

$$\beta^{(2,1)} = M^{-1}\beta^{(1,0)}\overline{w^{(0)}g^{(3)}}. \quad (4.4)$$

The pattern of the solutions is now clear and the details may be found in Chatwin's (1970) paper. To record the results, $f^{(3)}$ and $f^{(4)}$ have the forms

$$f^{(3)}(X) = \{\beta^{(3,0)}\mathrm{He}_2(X) + \beta^{(3,1)}\mathrm{He}_4(X) + \beta^{(3,2)}\mathrm{He}_6(X)\}\exp(-\tfrac{1}{2}X^2), \quad (4.5)$$

$$C \sim T^{-1}C^{(1)}+T^{-2}C^{(2)}+\ldots+T^{-n}C^{(n)}+\ldots$$

$$\begin{aligned}
C^{(1)} &= \beta^{(1,0)}H_0E \\
C^{(2)} &= \{\beta^{(2,0)}-\beta^{(1,0)}g^{(2)}\}\,H_1E+\beta^{(2,1)}H_3E \\
C^{(3)} &= \{\beta^{(3,0)}-\beta^{(2,0)}g^{(2)}+\beta^{(1,0)}g^{(3)}\}\,H_2E+\{\beta^{(3,1)}-\beta^{(2,1)}g^{(2)}\}\,H_4E+\beta^{(3,2)}H_6E \\
C^{(4)} &= -\beta^{(1,0)}g^{(4,2)}H_1E+\{\beta^{(4,0)}-\beta^{(3,0)}g^{(2)}+\beta^{(2,0)}g^{(3)}-\beta^{(1,0)}g^{(4,1)}\}\,H_3E \\
&\quad +\{\beta^{(4,1)}-\beta^{(3,1)}g^{(2)}+\beta^{(2,1)}g^{(3)}\}\,H_5E+\{\beta^{(4,2)}-\beta^{(3,2)}g^{(2)}\}\,H_7E+\beta^{(4,3)}\,H_9E \\
C^{(5)} &= -\beta^{(1,0)}g^{(5,3)}\,H_1E+\{\beta^{(1,0)}g^{(5,2)}-\beta^{(2,0)}g^{(4,2)}\}\,H_2E-\beta^{(2,1)}g^{(4,2)}\,H_4E \\
&\quad +\{\beta^{(5,0)}-\beta^{(4,0)}g^{(2)}+\beta^{(3,0)}g^{(3)}-\beta^{(2,0)}g^{(4,1)}+\beta^{(1,0)}g^{(5,1)}\}\,H_4E \\
&\quad +\{\beta^{(5,1)}-\beta^{(4,1)}g^{(2)}+\beta^{(3,1)}g^{(3)}-\beta^{(2,1)}g^{(4,1)}\}\,H_6E \\
&\quad +\{\beta^{(5,2)}-\beta^{(4,2)}g^{(2)}+\beta^{(3,2)}g^{(3)}\}\,H_8E+\{\beta^{(5,3)}-\beta^{(4,3)}g^{(2)}\}\,H_{10}E \\
&\quad +\beta^{(5,4)}\,H_{12}E+\psi_p^{(5)}
\end{aligned}$$

TABLE 2. The coefficients in the asymptotic series for the concentration; H_i and E denote $\mathrm{He}_i(X)$ and $\exp(-\frac{1}{2}X^2)$ respectively. The constants $\beta^{(r,s)}$ for $r > s \geqslant 1$ are linear functions of the $\beta^{(r,0)}$, and the term $\psi_p^{(5)}$ is defined by (4.8). Higher-order coefficients $C^{(j)}$, $j \geqslant 6$, are composed similarly of Hermite polynomials and particular solutions $\psi_p^{(j)}$, $j \geqslant 6$. The above coefficients differ from those found by Chatwin (1970) for a passive marker by the presence of terms containing the functions $g^{(r,s)}(r, \theta, X)$, $r \geqslant 4$, $s \geqslant 2$, and also by the particular solution $\psi_p^{(5)}$. These terms are caused by buoyancy effects at large times.

$$f^{(4)}(X) = \{\beta^{(4,0)}\mathrm{He}_3(X)+\beta^{(4,1)}\mathrm{He}_5(X)+\beta^{(4,2)}\mathrm{He}_7(X) + \beta^{(4,3)}\mathrm{He}_9(X)\}\exp(-\tfrac{1}{2}X^2), \qquad (4.6)$$

where the constants $\beta^{(r,s)}$, $r > s \geqslant 1$, are known linear functions of the $\beta^{(r,0)}$, $r \geqslant 1$, and are given in Chatwin's (1970) paper.

The equation for $f^{(5)}(X)$ reveals the first difference between the series for $\bar{C}$ in the cases of buoyant and passive markers. Equation (3.30) has the solution

$$f^{(5)}(X) = \{\beta^{(5,0)}\mathrm{He}_4(X)+\beta^{(5,1)}\mathrm{He}_6(X)+\beta^{(5,2)}\mathrm{He}_8(X)+\beta^{(5,3)}\mathrm{He}_{10}(X) + \beta^{(5,4)}\mathrm{He}_{12}(X)\}\exp(-\tfrac{1}{2}X^2)+\psi_p^{(5)}, \qquad (4.7)$$

where $\psi_p^{(5)}$ is a particular solution of (3.30) arising through buoyancy effects and defined by the equation

$$\left\{\frac{d^2}{dX^2}+X\frac{d}{dX}+5\right\}\psi_p^{(5)} = \frac{2}{M}f_{XX}^{(1)}\left\{\overline{w^{(0)}g^{(6,4)}}+\frac{\sigma}{P}\,\overline{(w^{(2)}g^{(4,2)}}+\overline{w^{(4)}g^{(2)})}\right\} + \frac{2}{M}f_X^{(1)}\left\{\overline{w^{(0)}\frac{\partial g^{(6,4)}}{\partial X}}+\frac{\sigma}{P}\overline{w^{(2)}\frac{\partial g^{(4,2)}}{\partial X}}+\sigma\left(\overline{u^{(3)}\frac{\partial g^{(4,2)}}{\partial r}}+\overline{\frac{1}{r}v^{(3)}\frac{\partial g^{(4,2)}}{\partial \theta}}+\overline{u^{(5)}\frac{\partial g^{(2)}}{\partial r}}\right)\right\}. \qquad (4.8)$$

The preceding results can be combined to give the asymptotic form for $\bar{C}$ as shown in table 2.

It only remains to determine the $\beta^{(r,0)}$ to specify the approach of the cloud of solute to the asymptotic state. Since the constants $\beta^{(r,0)}$ are related directly to the asymptotic form of the moments of the distribution (as shown shortly), it would suffice to determine a representation for the moments at large times. Unfortunately, it is found that numerical methods are required to determine the asymptotic form of the moments of distributions of buoyant markers.

The integral moments of C defined by

$$\nu_n(t) = \iiint_{\text{whole pipe}} (z-z_g)^n\, Cr\,dr\,d\theta\,dz \Big/ \iiint_{\text{whole pipe}} Cr\,dr\,d\theta\,dz$$

can be determined from (1.1) using the following properties of the Hermite polynomials:

$$\int_{-\infty}^{\infty} \mathrm{He}_m(X)\,\mathrm{He}_n(X)\exp(-\tfrac{1}{2}X^2)\,dX = (2\pi)^{\frac{1}{2}}\,m!\,\delta_{mn}, \tag{4.9}$$

$$\int_{-\infty}^{\infty} X^m \mathrm{He}_n(X)\exp(-\tfrac{1}{2}X^2)\,dX = \begin{cases} 0 \quad \text{if} \quad m < n \quad \text{or} \quad m-n \quad \text{is odd,} \\ (2\pi)^{\frac{1}{2}}\,m!(m-n-1)(m-n-3)\dots 1/(m-n)! \\ \qquad\qquad \text{if} \quad m-n \quad \text{is even.} \end{cases} \tag{4.10}$$

Using these properties, term-by-term integration of (1.1) gives

$$\iiint_{\text{whole pipe}} Cr\,dr\,d\theta\,dz \sim SPa\left\{(2\pi)^{\frac{1}{2}}\beta^{(1,0)} + \sum_{j=5}^{\infty} T^{1-j}\int_{-\infty}^{\infty}\psi_p^{(j)}\,dX\right\}, \tag{4.11}$$

where S denotes the cross-sectional area of the pipe. Now it is shown in §5 that

$$\int_{-\infty}^{\infty}\psi_p^{(5)}\,dX$$

is zero and the higher terms

$$\int_{-\infty}^{\infty}\psi_p^{(j)}\,dX, \quad j > 5,$$

in (4.11) should automatically be zero if the model equations are consistent. If this is assumed to be so, (4.11) reduces to

$$\iiint_{\text{whole pipe}} Cr\,dr\,d\theta\,dz = SPa(2\pi)^{\frac{1}{2}}\beta^{(1,0)}, \tag{4.12}$$

that is, the constant $\beta^{(1,0)}$ is proportional to the total amount of solute.

The z co-ordinate z_g of the centre of mass of the cloud of solute is given by

$$z_g = \iiint_{\text{whole pipe}} zCr\,dr\,d\theta\,dz \Big/ \iiint_{\text{whole pipe}} Cr\,dr\,d\theta\,dz$$

$$\sim Pa\left\{\beta^{(2,0)}(2\pi)^{\frac{1}{2}} + \sum_{j=6}^{\infty} T^{2-j}\int_{-\infty}^{\infty} X\psi_p^{(j)}\,dX\right\}(\beta^{(1,0)}(2\pi)^{\frac{1}{2}})^{-1}, \tag{4.13}$$

using the property (see §5)

$$\int_{-\infty}^{\infty} X\psi_p^{(5)}\,dX = 0.$$

This expression implies that the z co-ordinate of the centre of mass is constant, apart from some small, and possibly zero, terms induced by buoyancy effects at large times. Now, it is possible (and convenient) to choose the origin of the moving co-ordinate frame (r, θ, z) such that the constant $\beta^{(2,0)}$ in (4.13) is zero, that is,

$$\beta^{(2,0)} = 0$$

and

$$z_g \sim Pa(\beta^{(1,0)}(2\pi)^{\frac{1}{2}})^{-1}\sum_{j=6}^{\infty} T^{2-j}\int_{-\infty}^{\infty} X\psi_p^{(j)}\,dX. \tag{4.14}$$

Using the above results, it is found that the second moment $\nu_2(t)$ has the asymptotic form

$$\nu_2(t) \sim (Pa)^2\left\{T^2+2\frac{\beta^{(3,0)}}{\beta^{(1,0)}}+(\beta^{(1,0)}(2\pi)^{\frac{1}{2}})^{-1}T^{-2}\int_{-\infty}^{\infty}X^2\psi_p^{(5)}\,dX+O(T^{-3})\right\}. \quad (4.15)$$

The first two terms in this expression have the same form as the corresponding terms found by Chatwin (1970) for a passive marker, although $\beta^{(3,0)}$ now depends on buoyancy effects and is to be determined by matching (4.15) with the transient form of $\nu_2(t)$. The $O(T^{-2})$ term in (4.15) is caused by density currents at asymptotically large times and this term is interpreted in § 5.

It is now possible to compare the asymptotic expressions for $\nu_2(t)$ predicted by the present theory and by Erdogan & Chatwin's (1967) results. The term T^2 in (4.15) is given, from (1.2), by

$$T^2 = Mt\kappa/a^2$$

and since the constant M defined by (3.18) may be calculated as

$$M = 2/P^2+\tfrac{1}{24}, \quad (4.16)$$

T^2 may be approximated by $t\kappa/24a^2$ for flows in which P is large. ($P = Wa/\kappa$ is large when the convective speed W is great compared with typical cross-sectional diffusive speeds.) In these cases, (4.15) predicts

$$\nu_2(t) \sim \frac{a^2W^2}{24\kappa}t+2(Pa)^2\frac{\beta^{(3,0)}}{\beta^{(1,0)}}+\frac{(Pa)^2}{(2\pi)^{\frac{1}{2}}\beta^{(1,0)}}T^{-2}\int_{-\infty}^{\infty}X^2\psi_p^{(5)}\,dX+O(T^{-3}), \quad (4.17)$$

and the similarity between this equation and equation (4.6) of part 1 is obvious. The main conclusion of part 1 is therefore confirmed by this study; that is, *the dispersion induced by buoyancy effects at transient times* (this quantity is now represented by $2(Pa)^2\beta^{(3,0)}/\beta^{(1,0)}$) *is of greater order than the dispersion which is induced at asymptotically large times.*

The third moment of a distribution of buoyant solute is found to have the form

$$\nu_3(t) \sim (Pa)^3\left\{6T^2\frac{\beta^{(2,1)}}{\beta^{(1,0)}}+6\frac{\beta^{(4,0)}}{\beta^{(1,0)}}+O(T^{-2})\right\}, \quad (4.18)$$

where the $O(T^{-2})$ terms include the contributions to skewness of large-time buoyancy effects. Again, $\beta^{(4,0)}$ is to be determined by matching (4.18) with an intermediate-time representation of $\nu_3(t)$ and is dependent on transient buoyancy effects.

It follows from the above analysis that the properties of the moments and the character of the distribution may be determined from each other. To discuss the moments, take the Fourier–Laplace transform of the concentration equation [equation (2.4) in part 1] to obtain

$$-k^2a^2C^{\dagger\dagger}+\nabla^2C^{\dagger\dagger} = \frac{a^2}{\kappa}(pC^{\dagger\dagger}-\mathscr{C}^{(0)\dagger})+Pa\left(w\frac{\partial C}{\partial z}\right)^{\dagger\dagger}+\sigma\left(u\frac{\partial C}{\partial r}\right)^{\dagger\dagger}+\frac{\sigma}{r}\left(v\frac{\partial C}{\partial \theta}\right)^{\dagger\dagger}, \quad (4.19)$$

in which the velocity components and r have been non-dimensionalized as in § 2, a double dagger denotes a Fourier–Laplace transform and $\mathscr{C}^{(0)\dagger}$ is the Fourier transform of the initial distribution of solute. Since we may substitute

$$\begin{aligned} C^{\dagger\dagger}(k,r,\theta;p) &= \left(\frac{1}{2\pi}\right)^{\frac{1}{2}}\int_{-\infty}^{\infty} e^{ikz}\left\{\int_0^{\infty} e^{-pt}C(z,r,\theta;t)\,dt\right\}dz \\ &= \left(\frac{1}{2\pi}\right)^{\frac{1}{2}}\sum_{n=0}^{\infty}\frac{(ik)^n}{n\,!}\int_{-\infty}^{\infty} z^n C^{\dagger}(z,r,\theta;p)\,dz \\ &= \left(\frac{1}{2\pi}\right)^{\frac{1}{2}}\sum_{n=0}^{\infty}\frac{(ik)^n}{n\,!}C_n^{\dagger} \end{aligned}$$

for $C^{\dagger\dagger}$ and similar expressions for the other terms, the coefficients of $(ik)^n$ in the appropriately modified form of (4.19) would give an equation for the Laplace transform of the nth moment of the distribution. For passive markers, these moment equations are linear and have been examined by Aris (1956) and Chatwin (1970) to obtain asymptotic forms for the moments. This information enabled Chatwin to determine the unknown constants in the series for $\bar{C}(X,T)$. Unfortunately, for buoyant solutes, the moment equations involve transforms of nonlinear terms and can only be solved numerically. Such a numerical solution for the constants $\beta^{(r,0)}$ is beyond the scope of this paper so this gives the first conclusion mentioned in §1: the inability of analytical methods to predict the dispersion of buoyant solutes quantitatively.

5. The dispersion induced by buoyancy effects at large times

The term

$$T^{-2}\mathscr{D} = (Pa)^2\left((2\pi)^{\frac{1}{2}}\beta^{(1,0)}\right)^{-1}T^{-2}\int_{-\infty}^{\infty} X^2\psi_p^{(5)}\,dX \tag{5.1}$$

in (4.15) is now calculated to obtain the extra dispersion induced by large-time buoyancy effects. The computation of the cross-sectional means in (4.8) gives

$$\left\{\frac{d^2}{dX^2}+X\frac{d}{dX}+5\right\}\psi_p^{(5)} = \mathscr{R}, \tag{5.2}$$

where

$$\mathscr{R} = \frac{2}{M}f_{XX}^{(1)}\left(\alpha R f_X^{(1)}\right)^2 \times\left\{-\frac{21}{576\times 80P^4}+\frac{1}{P^2}O\left(\frac{1}{\sigma}\right)+\frac{46242}{100(576)^3\,1056}+O\left(\frac{1}{\sigma}\right)\right\}. \tag{5.3}$$ †

It may be verified from (4.2), (5.2) and (5.3) that

$$\int_{-\infty}^{\infty}\psi_p^{(5)}\,dX,\quad \int_{-\infty}^{\infty}X\psi_p^{(5)}\,dX$$

† The terms denoted by $O(\sigma^{-1})$ in this section are merely terms with coefficient σ^{-1} or σ^{-2}. An asymptotic theory based on the smallness of σ^{-1} is not implied, rather that these terms are negligible in the common cases where $\sigma = \nu/\kappa$ is large. (For example, σ had the value 2900 in the experiments of Reejhsinghani, Gill & Barduhn (1966) using du Pont's Pontamine 6BX dye in distilled water.) The terms denoted by $O(\sigma^{-1})$ come either from nonlinear terms in the governing equations or from unimportant terms which are not uniform axially.

are both zero, as stated in §4. Another simple calculation then shows that

$$\int_{-\infty}^{\infty} X^2 \psi_p^{(5)} dX = \frac{1}{2}\int_{-\infty}^{\infty} X^2 \mathscr{R} dX, \tag{5.4}$$

and the evaluation of this integral using (4.2) and (5.3) finally establishes the term $T^{-2}\mathscr{D}$ in (4.15).

Combining the above results, including (4.16) in particular, the time-dependent theory of part 2 predicts for the asymptotic form of $\nu_2(t)$

$$\nu_2(t) \sim t\left(\frac{a^2W^2}{24\kappa}+2\kappa\right)+2(Pa)^2\frac{\beta^{(3,0)}}{\beta^{(1,0)}}+\frac{a^2}{Mt\kappa}(\beta^{(1,0)})^2(a\alpha R)^2 Q_1^* + O(t^{-\frac{3}{2}}) \quad \text{as} \quad t\to\infty, \tag{5.5}$$

where Q_1^* is found to be

$$Q_1^* = \frac{1}{3\sqrt{3}}\frac{1}{48+P^2}\left(\frac{1}{2880}\right)^2\left[-60480+P^2 O\left(\frac{1}{\sigma}\right)+P^4\left(\frac{2569}{8448}+O\left(\frac{1}{\sigma}\right)\right)\right]. \tag{5.6}$$

In contrast, the expression for $\nu_2(t)$ obtained in part 1 was

$$\nu_2(t) \sim \frac{a^2W^2}{24\kappa}t + \text{constant} + \frac{a^2}{M^*t\kappa}\beta^2(a\alpha R)^2 Q_2^* \quad \text{as} \quad t\to\infty, \tag{5.7}$$

with Q_2^* defined by

$$Q_2^* = \frac{1}{3\sqrt{3}}\frac{1}{P^2}\left(\frac{1}{2880}\right)^2\left[-60480+P^4\frac{2569}{8448}+O\left(\frac{1}{\sigma}\right)\right]. \tag{5.8}$$

The constants M and M^* are given by

$$\tfrac{1}{2}M = P^{-2}+\tfrac{1}{48}, \quad \tfrac{1}{2}M^* = \tfrac{1}{48}. \tag{5.9}$$

The expressions (5.5) and (5.7) are similar qualitatively (and quantitatively for large σ and P). For large values of P, both Q_1^* and Q_2^* are positive and both become negative for P less than a critical Péclet number (which is approximately the same for both cases if σ is large). If the $O(\sigma^{-1})$ terms in (5.6) are neglected, Q_1^* changes sign at the critical Péclet number

$$P_c \approx 21{\cdot}12, \tag{5.10}$$

and in the experiments of Reejhsinghani *et al.* (1966), this Péclet number corresponds to an approximate mean flow speed of 10^{-3} cm/s in a tube of 1·5 mm internal diameter.

Clearly, the expression (5.7) is inaccurate for low values of the Péclet number since the $O(t^{-1})$ terms become infinite as $P\to 0$. The omission of the term $2\kappa t$ in (5.7) is not important since, as Erdogan & Chatwin (1967) remarked, it can be obtained easily by including axial molecular diffusion in a steady asymptotic theory. The expression (5.5) for $\nu_2(t)$ as predicted by part 2 is consistent at all Péclet numbers, including those near zero.

It is now possible to make some comments on the physical assumptions underlying Erdogan & Chatwin's (1967) model (see §2 of part 1). The time-dependent theory presented in part 2 demonstrates that Erdogan & Chatwin's arguments are sound provided that the Péclet and Schmidt numbers of the flow

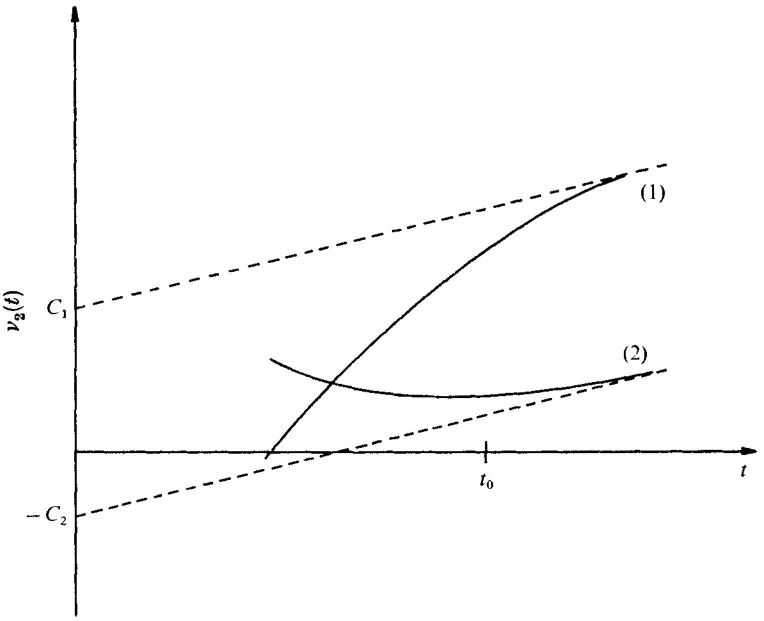

FIGURE 1. A schematic representation of the asymptotic form of $\nu_2(t)$. Curve (1) is $\nu_2(t) \sim t(a^2W^2/24\kappa+2\kappa)+C_1-\gamma_1/t$ and is the asymptote of $\nu_2(t)$ for $P_1 < P_c$. Line (2) is $\nu_2(t) \sim t(a^2W^2/24\kappa+2\kappa)-C_2+\gamma_2/t$ and is the asymptote of $\nu_2(t)$ for $P_2 > P_c$. The asymptotic theory is useful only for large t, e.g. $t > t_0$ (see text).

are both large. Their theory should be remarkably accurate for many solutes dispersing in liquids, although perhaps not for the dispersion of a buoyant contaminant in a laminar flow of gas. In brief, Erdogan & Chatwin selected for their model exactly those terms which are most important in describing the dispersion of buoyant solutes in solvents.

It seems worthwhile to describe the behaviour of $\nu_2(t)$ at large times by sketching it at two representative values of the Péclet number P. First consider dispersion when the Péclet number P_1 is small, with $0 < P_1 < P_c$. For small P, both Reejhsinghani *et al.* (1966) and Erdogan & Chatwin (1967) argue that mean axial spreading due to density currents is of more significance to the dispersion process than increased cross-sectional mixing (which reduces the dispersion). If this is true even for transient times, the constant $2(Pa)^2\beta^{(3,0)}/\beta^{(1,0)}$ in (5.5) should have a positive value, say, c_1, when $P_1 < P_c$. Also the $O(t^{-1})$ term is negative for $0 < P_1 < P_c$ and equal to $-\gamma_1/t$, say. In the second instance, consider a flow with a Péclet number P_2 greater than P_c. Then the arguments of the last-cited authors suggest that $2(Pa)^2\beta^{(3,0)}/\beta^{(1,0)}$ is negative and equal to $-c_2$, say, whilst the $O(t^{-1})$ term in (5.5) is positive and equal to γ_2/t. A schematic representation of $\nu_2(t)$ for these two situations is given in figure 1.

It is difficult to estimate the range of times for which the present theory is applicable. Chatwin (1970), in his study of dispersing passive solutes in Poiseuille flow, suggested that an expansion of the form (5.5) represented $\nu_2(t)$ to greater than 95% accuracy when $t > t_0$, where the non-dimensional time $\kappa t_0/a^2$ is 0·25 approximately. Presumably a similar range of validity holds for the case of buoyant solutes.

In conclusion, one further comparison may be made between parts 1 and 2. The coefficient functions $C^{(j)}$ given in table 2 show that buoyancy effects first modify the profile for $C(r, \theta, X)$ in the $O(T^{-4})$ terms, and the profile for $\bar{C}(X)$ in the term $T^{-5}\psi_p^{(5)}(X)$. This term satisfies an equation which is very similar to the equation (3.10) in part 1 for $h(X)$. Thus the theories predict a similar density modification to the profile $\bar{C}$.

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Appendix

A1. *The coefficients $u^{(n)}$ and $v^{(n)}$, $n \geqslant 3$*

A stream-function approach is inappropriate for terms $O(T^{-n})$ for $n \geqslant 3$, and the first instance of this failure may be seen by considering (2.25) with $n = 3$. [In this case, the term $w_X^{(2)}$ is known from (3.15) to be non-zero, and so (2.25) cannot be satisfied using a function with properties analogous to (3.3).] To avoid this problem, one of the velocity components $u^{(n)}$ and $v^{(n)}$ may be eliminated from (2.21), (2.22) and (2.25) in favour of the other to obtain a single equation with known inhomogeneous terms. For example, if $v^{(n)}$ is eliminated, the equation for $u^{(n)}$ takes the form

$$\left\{r^4\frac{\partial^4}{\partial r^4} + 6r^3\frac{\partial^3}{\partial r^3} + r^2\left(2\frac{\partial^4}{\partial\theta^2\,\partial r^2} + 5\frac{\partial^2}{\partial r^2}\right) + r\left(2\frac{\partial^3}{\partial\theta^2\,\partial r} - \frac{\partial}{\partial r}\right) + \frac{\partial^4}{\partial\theta^4} + 2\frac{\partial^2}{\partial\theta^2} + 1\right\}u^{(n)}$$

$$= \frac{r^2}{\sigma}\left\{\frac{\partial^2 A}{\partial\theta^2} - \frac{\partial^2}{\partial r\,\partial\theta}(rB)\right\} + \frac{r^2}{P}\left\{-\frac{2}{r}\frac{\partial^2}{\partial\theta^2} - \frac{\partial^2}{\partial r^2}\left(r\frac{\partial}{\partial r}(r)\right) - \frac{\partial^3}{\partial r\,\partial\theta^2} + \frac{\partial}{\partial r}\right\}w_X^{(n-1)},$$

where A and B are defined by

$$A = -\tfrac{1}{2}M(n-2)\,u^{(n-2)} - \tfrac{1}{2}MXu_X^{(n-2)} + w^{(0)}u_X^{(n-1)} + \frac{\sigma}{P}\sum_{j=1}^{n-2} w^{(j)}u_X^{(n-1-j)} - \frac{\sigma}{P^2}u_{XX}^{(n-2}$$

$$+\sigma\sum_{j=1}^{n-1}\left(u^{(j)}u_r^{(n-j)} + \frac{1}{r}v^{(j)}u_\theta^{(n-j)} - \frac{1}{r}v^{(j)}v^{(n-j)}\right) + R\alpha C^{(n)}\cos\theta$$

and

$$B = -\tfrac{1}{2}M(n-2)\,v^{(n-2)} - \tfrac{1}{2}MXv_X^{(n-2)} + w^{(0)}v_X^{(n-1)} + \frac{\sigma}{P}\sum_{j=1}^{n-2} w^{(j)}v_X^{(n-1-j)} - \frac{\sigma}{P^2}v_{XX}^{(n-2)}$$

$$+\sigma\sum_{j=1}^{n-1}\left(u^{(j)}v_r^{(n-j)} + \frac{1}{r}v^{(j)}v_\theta^{(n-j)} + \frac{1}{r}u^{(j)}v^{(n-j)}\right) - R\alpha C^{(n)}\sin\theta.$$

This equation may be solved for $u^{(n)}$ and the continuity equation (2.25) then gives $v^{(n)}$.

A2. *The important functions in table* 1

Some of the functions mentioned in table 1 are listed below. The remaining functions are unimportant since they lead to zero cross-sectional means in the equation for $\psi_p^{(5)}$ (see §A4).

$$k_1(r) = -\frac{\alpha R}{\sigma} f_X^{(1)} \frac{1}{2304}(r^6-6r^4+9r^2-4),$$

$$k_2(r) = \frac{\alpha R}{\sigma}\left\{\frac{1}{2304} f_X^{(2)}[-r^6+6r^4-9r^2+4] + \frac{1}{192P^2} f_{XX}^{(1)}[5r^4-10r^2+5] + \frac{1}{2764800} f_{XX}^{(1)}\left[\left(9+\frac{10}{\sigma}\right)r^{10} - \left(75+\frac{75}{\sigma}\right)r^8 + \left(250+\frac{150}{\sigma}\right)r^6 - \left(450+\frac{50}{\sigma}\right)r^4 + \left(405-\frac{100}{\sigma}\right)r^2 - \left(139-\frac{65}{\sigma}\right)\right]\right\},$$

$$k_5(r) = \left(\frac{\alpha R}{\sigma}\right)^2 f_X^{(1)} f_{XX}^{(1)}\left\{\frac{1}{737280}\frac{1}{P^2}\left(-\frac{1}{3}r^{11}+\frac{7}{2}r^9-\frac{25}{2}r^7+\frac{65}{3}r^5-20r^3+\frac{23}{3}r\right) + \frac{1}{\sigma}\frac{1}{179200(576)^2}\left(\frac{35}{18}r^{17}-40r^{15}+330r^{13}-\frac{4396}{3}r^{11}+3983r^9-6930r^7 + \frac{23170}{3}r^5-5320r^3+\frac{30907}{18}r\right) + \left(\frac{11}{576}\frac{1}{3840P^2}+\frac{1541}{(576)^2\,403200}\frac{1}{\sigma}\right)\tfrac{1}{2}(r^3-r)\right\},$$

$$l_1(r) = \frac{\alpha R}{\sigma} f_X^{(1)} \frac{1}{2304}(7r^6-30r^4+27r^2-4),$$

$$l_2(r) = \frac{\alpha R}{\sigma}\left\{\frac{1}{2304} f_X^{(2)}[7r^6-30r^4+27r^2-4] + \frac{1}{192P^2} f_{XX}^{(1)}[-r^4+6r^2-5] + \frac{1}{2764800} f_{XX}^{(1)}\left[-\left(99+\frac{170}{\sigma}\right)r^{10} + \left(675+\frac{1275}{\sigma}\right)r^8 - \left(1750+\frac{2850}{\sigma}\right)r^6 + \left(2250+\frac{2650}{\sigma}\right)r^4 - \left(1215+\frac{840}{\sigma}\right)r^2 + \left(139-\frac{65}{\sigma}\right)\right]\right\},$$

$$m_1(r) = \alpha R\frac{P}{\sigma} f_X^{(1)}\left[\frac{1}{8P^2}(-r^3+r) + \frac{1}{46080}\frac{1}{\sigma}(r^9-10r^7+30r^5-40r^3+19r)\right],$$

$$m_3(r) = -(\alpha R f_X^{(1)})^2\frac{P}{\sigma^2}\left\{\frac{1}{1474560P^2}[-4r^{10}+35r^8-100r^6+130r^4-80r^2+19] + \frac{1}{\sigma(576)^2\,358400}[35r^{16}-640r^{14}+4620r^{12}-17584r^{10}+39830r^8-55440r^6 + 46340r^4-21280r^2+4119] + \left(\frac{11}{576\times 3840P^2}+\frac{1541}{(576)^2\,403200\sigma}\right)(r^2-1)\right\}.$$

$\pi^{(3)}(X)$ satisfies

$$\frac{d\pi^{(3)}}{dX} = -(\alpha R f_X^{(1)})^2\frac{P^2}{\sigma^2}\left\{\frac{11}{576\times 960P^2}+\frac{1541}{(576)^2\,10080\sigma}\right\}.$$

A3. *The functions* $g^{(r,s)}(r,\theta,X), r \geqslant 4, s \geqslant 2$

The functions $g^{(4,1)}$ and $g^{(4,2)}$ are defined by (3.25) and (3.26). The functions $g^{(5,1)}$, $g^{(5,2)}$ and $g^{(5,3)}$ referred to in (3.27) are defined by the problems

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial g^{(5,1)}}{\partial r}\right) = w^{(0)}g^{(4,1)} - \overline{w^{(0)}g^{(4,1)}} - g^{(3)}\overline{w^{(0)}g^{(2)}} - g^{(2)}\overline{w^{(0)}g^{(3)}},$$

$$\nabla^2 g^{(5,2)} = w^{(0)}g^{(4,2)} - \overline{w^{(0)}g^{(4,2)}} + \frac{\sigma}{P}(w^{(2)}g^{(2)} - \overline{w^{(2)}g^{(2)}}) + \sigma u^{(2)}\frac{dg^{(3)}}{dr},$$

$$\nabla^2 g^{(5,3)} = \frac{\sigma}{P}w^{(3)} + \sigma\left(u^{(3)}\frac{dg^{(2)}}{dr} - \overline{u^{(3)}\frac{dg^{(2)}}{dr}}\right) + w^{(0)}\frac{\partial g^{(4,2)}}{\partial X} - \overline{w^{(0)}\frac{\partial g^{(4,2)}}{\partial X}}.$$

Also, $C^{(6)}$ has the form

$$\begin{aligned} C^{(6)} = {} & g^{(6,1)}(r)f^{(1)}_{XXXXX} + g^{(6,2)}(r,\theta,X)f^{(1)}_{XXX} + g^{(6,3)}(r,\theta,X)f^{(1)}_{XX} + g^{(6,4)}(r,\theta,X)f^{(1)}_{X} \\ & + g^{(5,1)}(r)f^{(2)}_{XXXX} + g^{(5,2)}(r,\theta,X)f^{(2)}_{XX} + g^{(5,3)}(r,\theta,X)f^{(2)}_{X} \\ & + g^{(4,1)}(r)f^{(3)}_{XXX} + g^{(4,2)}(r,\theta,X)f^{(3)}_{X} + g^{(3)}f^{(4)}_{XX} + g^{(2)}f^{(5)} + f^{(6)}, \end{aligned}$$

where the functions $g^{(6,1)}, \ldots, g^{(6,4)}$ are defined by

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial g^{(6,1)}}{\partial r}\right) = w^{(0)}g^{(5,1)} - \overline{w^{(0)}g^{(5,1)}} - g^{(4,1)}\overline{w^{(0)}g^{(2)}} - g^{(3)}\overline{w^{(0)}g^{(3)}} - g^{(2)}\overline{w^{(0)}g^{(4,1)}},$$

$$\begin{aligned} \nabla^2 g^{(6,2)} = {} & w^{(0)}g^{(5,2)} - \overline{w^{(0)}g^{(5,2)}} + \frac{\sigma}{P}(w^{(2)}g^{(3)} - \overline{w^{(2)}g^{(3)}}) + \sigma\left(u^{(2)}\frac{\partial g^{(4,1)}}{\partial r} - \overline{u^{(2)}\frac{\partial g^{(4,1)}}{\partial r}}\right) \\ & - g^{(4,2)}\overline{w^{(0)}g^{(2)}} - g^{(2)}\left(\overline{w^{(0)}g^{(4,2)}} + \frac{\sigma}{P}\overline{w^{(2)}g^{(2)}}\right), \end{aligned}$$

$$\begin{aligned} \nabla^2 g^{(6,3)} = {} & w^{(0)}g^{(5,3)} - \overline{w^{(0)}g^{(5,3)}} + \frac{\sigma}{P}(w^{(3)}g^{(2)} - \overline{w^{(3)}g^{(2)}}) + w^{(0)}\frac{\partial g^{(5,2)}}{\partial X} - \overline{w^{(0)}\frac{\partial g^{(5,2)}}{\partial X}} - \frac{2}{P^2}\frac{\partial g^{(4,2)}}{\partial X} \\ & + \sigma\left(u^{(3)}\frac{dg^{(3)}}{dr} - \overline{u^{(3)}\frac{dg^{(3)}}{dr}}\right) - g^{(2)}\left(2\overline{w^{(0)}\frac{\partial g^{(4,2)}}{\partial X}} + \frac{\sigma}{P}\overline{\frac{\partial w^{(2)}}{\partial X}g^{(2)}} + \sigma\overline{u^{(3)}\frac{\partial g^{(2)}}{\partial r}}\right), \end{aligned}$$

$$\begin{aligned} \nabla^2 g^{(6,4)} = {} & \frac{\sigma}{P}w^{(4)} + w^{(0)}\frac{\partial g^{(5,3)}}{\partial X} - \overline{w^{(0)}\frac{\partial g^{(5,3)}}{\partial X}} - g^{(2)}\left(\overline{w^{(0)}\frac{\partial^2 g^{(4,2)}}{\partial X^2}} + \sigma\overline{\frac{\partial u^{(3)}}{\partial X}\frac{\partial g^{(2)}}{\partial r}}\right) \\ & + \sigma\left(u^{(2)}\frac{\partial g^{(4,2)}}{\partial r} - \overline{u^{(2)}\frac{\partial g^{(4,2)}}{\partial r}} + \frac{1}{r}v^{(2)}\frac{\partial g^{(4,2)}}{\partial \theta} - \overline{\frac{1}{r}v^{(2)}\frac{\partial g^{(4,2)}}{\partial \theta}} + u^{(4)}\frac{\partial g^{(2)}}{\partial r} - \overline{u^{(4)}\frac{\partial g^{(2)}}{\partial r}}\right) \\ & - \frac{1}{P^2}\frac{\partial^2 g^{(4,2)}}{\partial X^2} - Mg^{(4,2)} - \tfrac{1}{2}MX\frac{\partial g^{(4,2)}}{\partial X}. \end{aligned}$$

In these equations, the $g^{(r,1)}$ have the same form as the corresponding functions found by Chatwin (1970) for a passive marker, and only $g^{(4,2)}$ and $g^{(6,4)}$ affect the equation for $\psi^{(5)}_p$ (see §A4). These functions have the form

$$\begin{aligned} g^{(4,2)}(r,\theta,X) = {} & \alpha R f^{(1)}_X \cos\theta\left[\frac{1}{5\,529\,600}\left\{\left(10+\frac{1}{\sigma}\right)r^{11} - \left(105+\frac{15}{\sigma}\right)r^9 + \left(375+\frac{75}{\sigma}\right)r^7\right.\right. \\ & \left.\left. - \left(650+\frac{200}{\sigma}\right)r^5 + \left(600+\frac{285}{\sigma}\right)r^3 - \left(340+\frac{256}{\sigma}\right)r\right\} + \frac{1}{192P^2}(-r^5+3r^3-4r)\right] \end{aligned}$$

and

$$
\begin{aligned}
g^{(6,4)}(r,\theta,X) = {} & (\alpha R f_X^{(1)})^2 \left[\frac{1}{2(576)^2 P^2} \left\{ r^{12}\left(\frac{1}{16}+\frac{1}{80\sigma}\right) - r^{10}\left(\frac{27}{40}+\frac{63}{400\sigma}\right) \right.\right. \\
& + r^8\left(\frac{93}{32}+\frac{45}{64\sigma}\right) - r^6\left(\frac{55}{8}+\frac{13}{8\sigma}\right) + r^4\left(9+\frac{327}{160\sigma}\right) - r^2\left(6+\frac{21}{16\sigma}\right) \\
& \left. + \left(\frac{139}{112}+\frac{2969}{11200\sigma}\right)\right\} + \frac{1}{32(576)^2}\left\{ -r^{18}\left(\frac{1}{4320}+\frac{1}{43\,200\sigma}+\frac{1}{1\,036\,800\sigma^2}\right)\right. \\
& + r^{16}\left(\frac{11}{2560}+\frac{7}{12\,800\sigma}+\frac{1}{4480\sigma^2}\right) - r^{14}\left(\frac{73}{2240}+\frac{29}{5600\sigma}+\frac{33}{15\,680\sigma^2}\right) \\
& + r^{12}\left(\frac{259}{1920}+\frac{263}{9600\sigma}+\frac{157}{14\,400\sigma^2}\right) - r^{10}\left(\frac{553}{1600}+\frac{37}{400\sigma}+\frac{569}{16\,000\sigma^2}\right) \\
& + r^8\left(\frac{1129}{1920}+\frac{2033}{9600\sigma}+\frac{99}{1280\sigma^2}\right) - r^6\left(\frac{251}{360}+\frac{4901}{14\,400\sigma}+\frac{331}{2880\sigma^2}\right) \\
& + r^4\left(\frac{91}{160}+\frac{287}{800\sigma}+\frac{22\,399}{201\,600\sigma^2}\right) - r^2\left(\frac{17}{60}+\frac{16}{75\sigma}+\frac{24\,743}{403\,200\sigma^2}\right) \\
& \left.\left. + \left(\frac{15\,343}{302\,400}+\frac{506\,917}{12\,096\,000\sigma}+\frac{5\,852\,377}{508\,032\,000\sigma^2}\right)\right\}\right] \\
& + \text{func.}\,(r)\cos\theta + \text{func.}\,(r)\cos 2\theta.
\end{aligned}
$$

A4. *The full equation for* $\psi_p^{(5)}$

The term $\psi_p^{(5)}(X)$ satisfies the equation

$$
\begin{aligned}
& \left(\frac{d^2}{dX^2}+X\frac{d}{dX}+5\right)\psi_p^{(5)} \\
& = \frac{2}{M} f_{XXX}^{(1)}\left[\overline{w^{(0)}g^{(6,2)}} + \frac{\sigma}{P}\overline{w^{(2)}g^{(4,1)}}\right] + \frac{2}{M} f_{XX}^{(1)}\left[\overline{w^{(0)}g^{(6,3)}} + \overline{w^{(0)}\frac{\partial g^{(6,2)}}{\partial X}}\right. \\
& \quad \left. + \frac{\sigma}{P}\overline{w^{(3)}g^{(3)}} + \sigma\overline{u^{(3)}\frac{\partial g^{(4,1)}}{\partial r}}\right] + \frac{2}{M} f_{X}^{(1)}\left[\overline{w^{(0)}g^{(6,4)}} + \overline{w^{(0)}\frac{\partial g^{(6,3)}}{\partial X}} + \frac{\sigma}{P}\left(\overline{w^{(2)}g^{(4,2)}} + \overline{w^{(4)}g^{(2)}}\right)\right. \\
& \quad \left. + \sigma\left(\overline{u^{(4)}\frac{\partial g^{(3)}}{\partial r}} + \left\{\overline{u^{(2)}\frac{\partial g^{(5,2)}}{\partial r}} + \overline{\frac{1}{r}v^{(2)}\frac{\partial g^{(5,2)}}{\partial \theta}}\right\}\right)\right] + \frac{2}{M} f^{(1)}\left[\overline{w^{(0)}\frac{\partial g^{(6,4)}}{\partial X}} + \frac{\sigma}{P}\overline{w^{(2)}\frac{\partial g^{(4,2)}}{\partial X}}\right. \\
& \quad \left. + \sigma\left(\overline{u^{(3)}\frac{\partial g^{(4,2)}}{\partial r}} + \overline{\frac{1}{r}v^{(3)}\frac{\partial g^{(4,2)}}{\partial \theta}} + \overline{u^{(5)}\frac{\partial g^{(2)}}{\partial r}} + \left\{\overline{u^{(2)}\frac{\partial g^{(5,3)}}{\partial r}} + \overline{\frac{1}{r}v^{(2)}\frac{\partial g^{(5,3)}}{\partial \theta}}\right\}\right)\right] \\
& \quad + \frac{2}{M} f_{XX}^{(2)}\left[\overline{w^{(0)}g^{(5,2)}} + \frac{\sigma}{P}\overline{w^{(2)}g^{(3)}}\right] + \frac{2}{M} f_{X}^{(2)}\left[\overline{w^{(0)}g^{(5,3)}} + \overline{w^{(0)}\frac{\partial g^{(5,2)}}{\partial X}} + \frac{\sigma}{P}\overline{w^{(3)}g^{(2)}} + \sigma\overline{u^{(3)}\frac{\partial g^{(3)}}{\partial r}}\right] \\
& \quad + \frac{2}{M} f^{(2)}\left[\overline{w^{(0)}\frac{\partial g^{(5,3)}}{\partial X}} + \sigma\left(\overline{u^{(4)}\frac{\partial g^{(2)}}{\partial r}} + \left\{\overline{u^{(2)}\frac{\partial g^{(4,2)}}{\partial r}} + \overline{\frac{1}{r}v^{(2)}\frac{\partial g^{(4,2)}}{\partial \theta}}\right\}\right)\right] \\
& \quad + \frac{2}{M} f_{X}^{(3)}\left[\overline{w^{(0)}g^{(4,2)}} + \frac{\sigma}{P}\overline{w^{(2)}g^{(2)}}\right] + \frac{2}{M} f^{(3)}\left[\overline{w^{(0)}\frac{\partial g^{(4,2)}}{\partial X}} + \sigma\overline{u^{(3)}\frac{\partial g^{(2)}}{\partial r}}\right].
\end{aligned}
$$

Here, many terms can be seen to vanish since they are the zero cross-sectional means of terms of the form func. $(r)\cos n\theta$. A simple argument based on the definition of the stream function $\psi^{(2)}(r, \theta, X)$ [(3.3) and (3.4)] shows that the terms included in the curly brackets give no contribution.

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