J. Fluid Mech. (1997), *vol.* 334, *pp.* 111–133

Unsteady heat or mass transport from a suspended particle at low Péclet numbers

By C. POZRIKIDIS

Department of Applied Mechanics and Engineering Sciences, University of California at San Diego, La Jolla, CA 92093-0411, USA

e-mail: costas@ames.ucsd.edu

(Received 21 September 1995 and in revised form 11 September 1996)

Unsteady heat or mass transport from a particle with an arbitrary shape suspended in a fluid of infinite expanse is considered in the limit of small Péclet numbers where diffusion is dominant. In a frame of reference in which the particle appears to be stationary, the velocity of the fluid is uniform or varies in a linear manner with respect to the spatial coordinates, with an arbitrary time dependence. The temperature or concentration of a species at the surface of the particle is held at a certain constant value, whereas that at infinity is held at another constant value. Two particular problems are considered, both to leading order with respect to the Péclet number: (*a*) the rate of transport from a particle that is introduced suddenly into a steady flow near the steady state; and (*b*) the average rate of transport from a particle that is suspended in a time-periodic flow. The theory uses the method of matched asymptotic expansions and employs the Green's function of the convection–diffusion equation for a generally unsteady uniform or linear flow. The Green's function is derived in closed form by first performing a transformation to a Lagrangian framework. In the first problem of transient transport, it is found that the functional form of the rate of transport near the steady state is affected strongly by the structure of the incident flow: the decay in uniform or elongational flow is exponential, whereas the decay in simple shear flow is algebraic. In the second problem of transport in a periodic flow, it is found that the value of a properly defined frequency parameter has a strong influence on the mean rate of transport, for all types of flow. The oscillation induces convective mixing and thereby reduces the mean rate of transport by a substantial factor. The ability of the theory to describe another situation of heat or mass transport considered by Pedley is also discussed.

1. Introduction

Consider a small particle with an arbitrary shape, suspended in a quiescent surrounding fluid of infinite expanse, and assume that the temperature or concentration of a certain species at the surface of the particle is held at the constant value T_1, while the temperature or concentration far from the particle has the lower value T_0. Heat or mass is released from the particle surface at a constant rate Q_0 that depends upon the particle shape alone. The distribution of the temperature or concentration field is governed by the steady diffusion equation whose linear nature, along with that of the boundary conditions, allows us to write $Q_0 = h_0(T_1 - T_0)$, where h_0 is a heat or mass transfer coefficient. It is convenient to express h_0 in terms of the Nusselt number $Nu_0 = h_0/4\pi ak = Q_0/4\pi ak(T_1 - T_0)$, where a is half the maximum particle diameter, and k is the thermal conductivity or species mass diffusivity. For a spherical particle of radius a, it is readily found that $Nu_0 = 1$.

When in a frame of reference in which the particle appears to be stationary the motion of the ambient fluid is steady, the rate of heat or mass transport from the particle changes by an amount $Q - Q_0$ that is a function of the structure and intensity of the incident flow in the vicinity of the particle, as well as of the value of the Péclet number $Pe = aU/D$; U is the typical magnitude of the velocity of the fluid in the vicinity of the particle, and D is the thermal or species mass diffusivity. In the case of heat transfer, $D = k/\rho c_p$, where ρ and c_p are the density and heat capacity of the fluid at constant pressure, and in the case of mass transfer $D = k$. Assuming that neither the temperature nor concentration field has a significant effect on the physical properties of the fluid, we write $Q = h(T_1 - T_0)$, with the understanding that the heat or mass transfer coefficient h, and its dimensionless version expressed by the Nusselt number $Nu = h/4\pi ak = Q/4\pi ak(T_1 - T_0)$, depend upon the structure and intensity of the ambient flow, as well as on the value of the Péclet number Pe.

The dependencies of Nu on the Péclet number and on the Reynolds number $Re = aU/\nu$, where ν is the kinematic viscosity of the fluid, have been studied on many occasions using the method of matched asymptotic expansions, for several types of flows (Acrivos & Taylor 1962; Brenner 1963; Frankel & Acrivos 1968; Clift, Grace & Weber 1978; Acrivos 1980; Brunn 1984). Batchelor (1979) extended and unified prior analyses in the limit of small and large values of the Péclet number. It is now well-established that, for small values and to leading order with respect to Pe, Nu is independent of Re but depends only on the structure of the incident flow.

The more general problem of unsteady transport from a particle that is suspended in a steady or unsteady incident flow has received much less attention, as will be discussed in the following sections. In this case, the distribution of the scalar field around the particle is governed by the unsteady convection–diffusion equation

$$\frac{\partial T}{\partial t} + \nabla \cdot (\boldsymbol{u}(t)\, T) = D\nabla^2 T \tag{1.1}$$

with boundary conditions $T = T_1$ on the particle surface, and $T = T_0$ far from the particle. The rate of scalar transport from the particle surface is a time-dependent function $Q(t)$ whose instantaneous value is affected not only by the current structure of the flow, but also by the history of the fluid and particle motion.

Close to the particle surface, for $r < r_c$, where r is the distance from a designated particle centre and r_c is a certain critical distance, convective effects are secondary and the distribution of the temperature or concentration is governed by the unsteady diffusion equation, subject to boundary conditions to be discussed in the following paragraphs. Far from the particle surface, for $r > r_c$, both convective and diffusive effects are significant, and the distribution of the temperature or concentration is governed by the full form of the unsteady convection–diffusion equation (1.1). A schematic illustration of these two regimes is depicted in figure 1.

If the velocity of translation of the particle is roughly equal to the velocity of the incident flow evaluated at the designated particle centre, with an error that is comparable to the magnitude of the local velocity gradient γ multiplied by the particle size a, then $r_c/a \approx Pe^{-1/2}$ where $Pe = \gamma a^2/D$. But if the particle velocity differs from the velocity of the fluid by an amount that is larger than γa, then $r_c/a \approx Pe^{-1}$ where $Pe = aU/D$ and U is the velocity of the fluid relative to that of the particle. When Pe is small, the distance r_c is much larger than the characteristic particle size a, and the distribution of the scalar in the outer regime is insensitive to the precise particle shape.

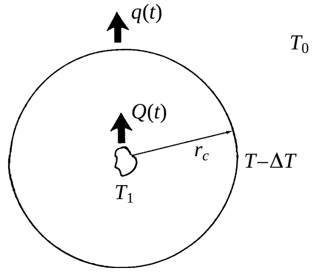

FIGURE 1. Unsteady transport from a suspended particle in a generally unsteady ambient flow at small Péclet numbers. Schematic illustration of the inner and outer transport regimes where convective transport is, respectively, negligible or significant.

To describe the temperature or concentration field in the outer regime, we work in a frame of reference in which the particle appears to be stationary, and define the rate of heat or mass transport $q(t)$ across a spherical surface that is centred at the particle and whose radius R is comparable to r_c. Then, following Batchelor (1979, p. 374) and previous authors, we approximate the actual fluid velocity $\boldsymbol{u}$ with the velocity of the unperturbed incident flow $\boldsymbol{u}^\infty(t)$. This approximation requires that the disturbance flow has decayed, the distortion of the velocity profile due to the establishment of a wake has virtually disappeared, and any steady streaming motion due to an oscillatory flow has ceased at the radial distance r_c. Hydrodynamic instabilities aside, these conditions will be met independently of the value of the Reynolds number, as long as the Péclet number is sufficiently small.

To leading order with respect to Pe, the distribution of the scalar in the outer regime may then be expressed in the form

$$T^{Outer}(\boldsymbol{x}, t; \boldsymbol{x}_0) = \alpha \int_{t_s}^{t} q(t_0)\, G^{CD}(\boldsymbol{x}, t; \boldsymbol{x}_0, t_0)\, \mathrm{d}t_0 + T_0, \tag{1.2}$$

where $\boldsymbol{x}_0$ is the designated particle centre which, in the aforementioned frame of reference is stationary, t_s is the time at which scalar transport has begun, $\alpha = D/k$, and the subscript s stands for *start*. For heat transport, $\alpha = 1/\rho c_p$, and for mass transport $\alpha = 1$. The kernel G^{CD}, with dimensions of inverse cubed length, is the Green's function of the convection–diffusion equation, defined as the solution of the equation

$$\frac{\partial G^{CD}}{\partial t} + \boldsymbol{\nabla}\cdot(\boldsymbol{u}^\infty(t)\, G^{CD}) = D\nabla^2 G^{CD} + \delta(x-x_0)\,\delta(y-y_0)\,\delta(z-z_0)\,\delta(t-t_0), \tag{1.3}$$

where δ is the one-dimensional delta function. Furthermore, G^{CD} is required to decay as $\boldsymbol{x}$ moves far away from $\boldsymbol{x}_0$.

To compute the rate of transport from the particle surface, we work in two steps. First, we take the limit of (1.2) as the field point $\boldsymbol{x}$ tends to the particle centre $\boldsymbol{x}_0$ and derive the singular asymptotic expansion

$$T^{Outer}(t) \to \alpha\, q(t) \frac{1}{4\pi D\, |\boldsymbol{x}-\boldsymbol{x}_0|} + T_0 - \Delta T(t) + \ldots, \tag{1.4}$$

where $G^{D\text{-}S} = 1/(4\pi|\boldsymbol{x}-\boldsymbol{x}_0|)$ is the Green's function of the steady diffusion equation that satisfies the equation $\nabla^2 G^{D\text{-}S} + \delta(x-x_0)\,\delta(y-y_0)\,\delta(z-z_0) = 0$. As $\boldsymbol{x}$ tends to $\boldsymbol{x}_0$, the terms represented by the dots tend to vanish. The first term on the right-hand side of (1.4) describes the quasi-steady scalar field that prevails in the absence of convection. The instantaneous value of the quantity $\Delta T(t)$ depends upon the variation of $q(t)$ over all previous times up to the present time, and it is generally a function of the direction of $\boldsymbol{x}-\boldsymbol{x}_0$. If the instantaneous velocity of the incident flow at the designated particle centre $\boldsymbol{x}_0$ vanishes, then $\Delta T(t)$ is independent of the direction of $\boldsymbol{x}-\boldsymbol{x}_0$. This can be seen by expanding $\boldsymbol{u}^\infty(t)$ in a Taylor series about the point $\boldsymbol{x}_0$, reraining the leading-order term, and then inspecting the functional form of the Green's function for uniform and linear flow discussed by Batchelor (1979).

Secondly, we consider the distribution of the scalar field in the inner diffusive regime, for $r < r_c$, and use the general principles of matched asymptotic expansions, to find that the boundary conditions accompanying the governing unsteady diffusion equation are (*a*) $T = T_1$ at the particle surface, and (*b*) $T = T_0 - \Delta T(t)$ at a radial distance R that is comparable to r_c, as shown in figure 1. Integrating the unsteady diffusion equation from the initial time t_s up to a certain time t allows us to express the rate of transport from the particle surface $Q(t)$ in terms of $\Delta T(t)$. A relation between the functions $q(t)$ and $\Delta T(t)$ emerges by identifying $q(t)$ with the rate of transport across a spherical surface of radius R, and this, in conjunction with (1.4) completes the mathematical formulation. Obtaining quantitative results, however, requires a more precise definition for the radius R.

When the distribution of the temperature or concentration field has reached a steady state, the absence of heat or mass generation or dissipation requires that $q(t) = Q(t)$. In this case, R may be consistently shifted to infinity to a leading-order approximation, and this permits an analytical or numerical computation (Batchelor 1979). There are at least two additional circumstances where a precise definition for R is not necessary in order to make further progress. The first one concerns the late stages of transport from a particle that has been introduced suddenly into a steady flow, near the steady state, to be discussed in §2. The second one concerns the mean rate of transport from a particle in a time-period flow, to be discussed in §3.

In §§2 and 3, we review and discuss the mathematical formulation for the aforementioned two problems, and in §§4 and 5 we derive analytical and numerical results for uniform incident flow and for a family of flows with a linear variation in space, with particular attention to simple shear flow and purely straining two-dimensional and axisymmetric flows. The derivation of quantitative results hinges upon our ability to compute the Green's function of the convection–diffusion for a generally unsteady flow with velocity field given by $\boldsymbol{u}^\infty(t) = \boldsymbol{A}(t)\cdot\boldsymbol{x} + \boldsymbol{U}(t)$, where $\boldsymbol{A}(t)$ is the uniform time-dependent velocity gradient and $\boldsymbol{U}(t)$ is a time-dependent velocity; this is discussed in the Appendix. In §6, we summarize the results, compare the present method of analysis with analogous methods of low-Reynolds-number hydrodynamics, and discuss the applicability to the theory to the problem of heat or mass transport from a small patch on an insulated or isothermal wall considered by Pedley (1972, 1976).

2. Late stages of transport in a steady flow

We consider heat or mass transport from a particle which, at time t_s, is immersed in an infinite fluid, and refer to a frame of reference in which the particle appears to be stationary and the incident flow is steady. The behaviour of the rates of transport from the particle surface and from the boundary of the inner regime, $Q(t)$ and $q(t)$, are illustrated schematically in figure 2.

Konopliv & Sparrow (1971, 1972), Choudhury & Drake (1971), Abramzon & Elata (1984), Feng & Michaelides (1996), and others (see Clift *et al.* 1978) studied the asymptotic behaviour of the temperature of concentration field from a *spherical*

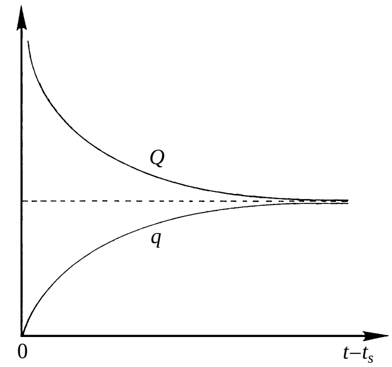

FIGURE 2. Behaviour of the rates of transport from the particle surface, $Q(t)$, and the rate of transport across the boundary that demarkates the inner diffusive regime from the outer convective–diffusive regime, $q(t)$, for a particle that is immersed suddenly in a steady flow.

particle immersed in a *uniform* flow at short and long elapsed times, for small and large values of the Péclet number. Polyanin (1984, p. 171) pointed out that the results for long times and small Péclet numbers are also applicable to arbitrary particle shapes. We are interested in evaluating the rate of transport from a particle of arbitrary shape immersed in a general, not necessarily uniform, incident flow.

A scaling analysis shows that when $t-t_s \gg r_c^2/D \approx (a/U)\,Pe^{-1}$ for uniform flow or $t-t_s \gg r_c^2/D \approx 1/\gamma$ for linear flow, the scalar field in the inner regime develops in a quasi-steady or parametric manner. Thus, the rate of heat or mass transport into the outer regime may be approximated by the instantaneous rate of transport from the particle surface, that is, $q(t) \approx Q(t)$. Following Batchelor (1979), we find that, accurate to leading order with respect to the Péclet number, the rate of transport from the particle surface is given by

$$Q(t) = h_0(T_1 - T_0 + \langle \Delta T(t) \rangle), \tag{2.1}$$

where the angle brackets designate the average value of $\Delta T(t)$ defined in (1.4) over all orientations of the vector $\boldsymbol{x}-\boldsymbol{x}_0$. The spatial fluctuations of the quantity $\Delta T(t)$ with respect to $\boldsymbol{x}-\boldsymbol{x}_0$ around its mean value give rise to a diffusive scalar field that does not contribute to the net rate of transport. Equation (2.1) may be rewritten in terms of the relative increase of the rate of transport due to the flow as

$$\frac{Q(t)-Q_0}{Q_0} = \frac{\langle \Delta T(t) \rangle}{T_1 - T_0} = Nu_0 \frac{4\pi a k \langle \Delta T(t) \rangle}{Q_0}, \tag{2.2}$$

where Q_0 is the purely diffusive steady rate of transport that is established at long times when the particle is introduced and remains stationary in a quiescent fluid. To compute the quantity $\langle \Delta T(t) \rangle$, we work in two stages.

First, we note that, because the flow is steady, the arguments t and t_0 of the Green's function G^{CD} defined in (1.3) combine to form their difference, and (1.2) takes the form

$$T^{Outer}(\boldsymbol{x}, t; \boldsymbol{x}_0) = \alpha \int_0^{t-t_s} q(t-\hat{t})\, G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0)\, \mathrm{d}\hat{t} + T_0, \tag{2.3}$$

where $\hat{t} = t - t_0$. As long as $t-t_s \gg r_c^2/D$, it is consistent to replace $q(t)$ with its asymptotic value $q(\infty) = Q(\infty)$. The results of the following two sections will show that this approximation introduces a relative error that is an algebraically or exponentially decaying function of $t-t_s$. A rigorous justification for this approximation can be made by working with the Laplace-transformed variables as discussed, for example, by Polyanin (1984, p. 171). Furthermore, since we are interested in evaluating the leading-order effect in the Péclet number, it is consistent to replace the actual value $Q(\infty)$ with the purely diffusive value Q_0. These simplifications transform (2.3) to

$$T^{Outer}(\boldsymbol{x}, t; \boldsymbol{x}_0) = \alpha\, Q_0 \int_0^{t-t_s} G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0)\, \mathrm{d}\hat{t} + T_0. \tag{2.4}$$

Secondly, we introduce the Green's function of the unsteady diffusion equation G^D, which satisfies the equation

$$\frac{\partial G^D}{\partial t} = D\nabla^2 G^D + \delta(x-x_0)\,\delta(y-y_0)\,\delta(z-z_0)\,\delta(t-t_0) \tag{2.5}$$

and vanishes at infinity, and note that the corresponding stationary Green's function $G^{D\text{-}S} = 1/(4\pi|\boldsymbol{x}-\boldsymbol{x}_0|)$ is

$$G^{D\text{-}S}(\boldsymbol{x}; \boldsymbol{x}_0) = \int_0^{\infty} G^D(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0)\, \mathrm{d}\hat{t}. \tag{2.6}$$

Rearranging the integral on the right-hand side of (2.4), and taking into account (2.6), we obtain

$$\begin{aligned} T^{Outer}(\boldsymbol{x}, t; \boldsymbol{x}_0) &= \alpha \frac{Q_0}{4\pi D\,|\boldsymbol{x}-\boldsymbol{x}_0|} \\ &\quad + T_0 - \alpha\, Q_0 \int_0^{\infty} (G^D(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0))\, \mathrm{d}\hat{t} - \alpha\, Q_0 \int_{t-t_s}^{\infty} G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0)\, \mathrm{d}\hat{t}. \end{aligned} \tag{2.7}$$

Finally, we take the limit of (2.7) as the field point $\boldsymbol{x}$ tends to the designated particle centre $\boldsymbol{x}_0$, and compare it with (1.4) to obtain

$$\begin{aligned} \Delta T(t) &= \lim_{\boldsymbol{x}\to\boldsymbol{x}_0} \left(\alpha\, Q_0 \int_0^{\infty} [G^D(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0)]\, \mathrm{d}\hat{t} \right) \\ &\quad + \alpha\, Q_0 \int_{t-t_s}^{\infty} G^{CD}(\boldsymbol{x}_0, \hat{t}; \boldsymbol{x}_0)\, \mathrm{d}\hat{t}. \end{aligned} \tag{2.8}$$

Unless the incident velocity vanishes at $\boldsymbol{x}_0$, the limit on the right-hand side of (2.8) may not be transferred into the integral. Substituting (2.8) into (2.2), we find

$$\frac{Q(t) - Q_0}{Q_0} = Nu_0\, F(t), \tag{2.9}$$

where
$$F(t) = \frac{4\pi a k \langle \Delta T(t) \rangle}{Q_0} = F(\infty) + 4\pi a D \int_{t-t_s}^{\infty} G^{CD}(\boldsymbol{x}_0, \hat{t}; \boldsymbol{x}_0)\, \mathrm{d}\hat{t} \tag{2.10}$$

and
$$\begin{aligned} F(\infty) &= 4\pi a D \lim_{\boldsymbol{x}\to\boldsymbol{x}_0} \left\langle \int_0^{\infty} \left(G^D(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0) \right) \mathrm{d}\hat{t} \right\rangle \\ &= \lim_{\boldsymbol{x}\to\boldsymbol{x}_0} \left\langle \frac{a}{|\boldsymbol{x}-\boldsymbol{x}_0|} - 4\pi a D G^{CD\text{-}S}(\boldsymbol{x}; \boldsymbol{x}_0) \right\rangle. \end{aligned} \tag{2.11}$$

The magnitude of the dimensionless constant $F(\infty)$, corresponding to the limit of steady transport, vanishes as the Péclet number tends to zero, but its precise leading-order dependence on Pe depends on the structure of the incident flow. The computation of this constant for uniform flow and for a family of flows varying linearly in space was discussed by Batchelor (1979) in terms of the integral on the right-hand side of (2.11).

Choudhury & Drake (1971), Polyanin (1984), and Feng & Michaelides (1996) computed the function $F(t)$ for uniform flow past a spherical or an arbitrary particle using the method of the Laplace transform. In §§4 and 5, we shall re-derive the results using the present theory and, in addition, we shall compute the asymptotic form of the function $F(t)$ for a family of linear flows in terms of the second integral on the right-hand side of (2.10). The results will demonstrate the importance of the structure of the flow for the manner in which the rate of transport approaches its asymptotic value near the steady state.

3. Transport in a periodic flow

Fluctuating flows are often used in chemical engineering processes of heat and mass transfer involving two-phase and particulate flow. The applied engineering literature contains a large number of relevant experimental investigations and accompanying empirical correlations, reviewed by Clift *et al.* (1978, pp. 312, 314). Owing to the intricacies of the mathematical problem, a comprehensive theory has not been established.

Batchelor (1980) considered the rate of transport from a spherical rigid particle that is suspended in a fluctuating turbulent flow at large values of the Péclet number, under the assumption that, in a frame of reference in which the particle is stationary, the flow occurs at low Reynolds numbers. We are interested in the diametrically opposite limit of transport at small Péclet numbers where the Reynolds number does not enter the analysis to a leading-order approximation.

We begin developing the theory by taking the time average of the unsteady diffusion equation over one period, indicated by an overbar, and note that the time-averaged temperature or concentration field $\bar{T}$ in the inner regime satisfies the steady diffusion equation and is thus amenable to the theory of Batchelor (1979). Furthermore, we expect that the quantity $\Delta T(t)$ defined in (1.4) will be a periodic function of time, and may thus be expressed as a Fourier series. Taking the average of both sides of (1.4) over one period, we find

$$\bar{T}^{Outer} \to \alpha\, \bar{q} \frac{1}{4\pi D|\boldsymbol{x}-\boldsymbol{x}_0|} + T_0 - \overline{\Delta T} + \dots . \tag{3.1}$$

Since neither heat nor mass accumulates within the fluid over a period, the net rate of transport across any surface enclosing the particle over one period is constant, and we may put $\bar{q} = \bar{Q}$. Noting that the fluctuating component of $\langle \Delta T \rangle$ does not contribute to the net rate of transport from the particle, and following the arguments of Batchelor (1979), we write

$$\bar{Q} = h_0(T_1 - T_0 + \langle \overline{\Delta T} \rangle) \tag{3.2}$$

which may be rearranged to give

$$\frac{\bar{Q} - Q_0}{Q_0} = \frac{\langle \overline{\Delta T} \rangle}{T_1 - T_0} = Nu_0 \frac{4\pi a k \langle \overline{\Delta T} \rangle}{Q_0}. \tag{3.3}$$

To compute the mean value of the quantity $\Delta T(t)$ over one period, we consider the scalar distribution in the outer regime given by (1.2), and replace the instantaneous rate of transport $q(t)$ within the integral with the purely diffusive rate of transport Q_0. This substitution is permissible as long as we confine our attention to the leading-order effect with respect to the Péclet number. We may now proceed in two seemingly different but essentially equivalent ways.

In the first method, we recast (1.2) with $q(t) = Q_0$ into the form

$$T^{Outer}(\boldsymbol{x}, t; \boldsymbol{x}_0) = \alpha Q_0 \frac{1}{4\pi D|\boldsymbol{x}-\boldsymbol{x}_0|} + T_0 - \alpha Q_0 \int_{-\infty}^{t} (G^D(\boldsymbol{x}, t-t_0; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, t; \boldsymbol{x}_0, t_0))\,\mathrm{d}t_0, \tag{3.4}$$

where G^D was defined in (2.5). Considering the integrand on the right-hand side, we note that the limit of $\boldsymbol{x}$ tends to $\boldsymbol{x}_0$ and t tends to t_0, G^D and G^{CD} exhibit identical singular behaviour. As a result, the second integral tends to obtain a finite value that generally depends upon the direction of $\boldsymbol{x}-\boldsymbol{x}_0$. If the velocity happens to vanish at the location of $\boldsymbol{x}_0$, the limiting value of the integral is independent of the direction of $\boldsymbol{x}-\boldsymbol{x}_0$ (e.g. Batchelor 1979). We thus obtain

$$\Delta T(t) = \lim_{\boldsymbol{x}\to\boldsymbol{x}_0} \left(\alpha Q_0 \int_{-\infty}^{t} (G^D(\boldsymbol{x}, t-t_0; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, t; \boldsymbol{x}_0, t_0))\,\mathrm{d}t_0 \right), \tag{3.5}$$

where, in general, the limit may not be transferred into the integral.

Another method of computing the quantity ΔT involves introducing the Green's function of the unsteady convection–diffusion equation, and the Green's function of the steady convection–diffusion equation, computed by freezing the velocity at its instantaneous value at the current time t, denoted respectively by $G^{CD}_{(t)}$ and $G^{CD\text{-}S}_{(t)}$. These are related by an equation that is analogous to (2.6). We then recast (1.2) with $q(t) = Q_0$ into the form

$$T^{Outer}(\boldsymbol{x}, t; \boldsymbol{x}_0) = \alpha Q_0 G^{CD\text{-}S}_{(t)}(\boldsymbol{x}; \boldsymbol{x}_0) + T_0 - \alpha Q_0 \int_{-\infty}^{t} (G^{CD}_{(t)}(\boldsymbol{x}, t-t_0; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, t; \boldsymbol{x}_0, t_0))\,\mathrm{d}t_0. \tag{3.6}$$

As $\boldsymbol{x}$ tends to $\boldsymbol{x}_0$, the limiting value of the first term on the right-hand side may depend upon the direction of $\boldsymbol{x}-\boldsymbol{x}_0$, but the limiting value of the integral is independent of the direction of $\boldsymbol{x}-\boldsymbol{x}_0$. We thus obtain

$$\Delta T(t) = \alpha Q_0 C(t) + \alpha Q_0 \int_{-\infty}^{t} (G^{CD}_{(t)}(\boldsymbol{x}_0, t-t_0; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}_0, t; \boldsymbol{x}_0, t_0))\,\mathrm{d}t_0, \tag{3.7}$$

where the value of the dimensional coefficient $C(t)$ generally depends on the direction of $\boldsymbol{x}-\boldsymbol{x}_0$, and may be deduced from the analysis of steady transport in a steady flow.

Substituting (3.5) and (3.7) into (3.3) we finally obtain

$$\frac{\bar{Q}-Q_0}{Q_0} = Nu_0\,\overline{\varPhi}, \tag{3.8}$$

where the function $\varPhi$ is given by the two equivalent expressions

$$\varPhi(t) = 4\pi a D \left\langle \lim_{\boldsymbol{x}\to\boldsymbol{x}_0} \left(\int_{-\infty}^{t} [G^D(\boldsymbol{x}, t-t_0; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}, t; \boldsymbol{x}_0, t)]\,\mathrm{d}t_0 \right) \right\rangle \tag{3.9}$$

and

$$\varPhi(t) = 4\pi a D \langle C(t) \rangle + 4\pi a D \int_{-\infty}^{t} (G^{CD}_{(t)}(\boldsymbol{x}_0, t-t_0; \boldsymbol{x}_0) - G^{CD}(\boldsymbol{x}_0, t; \boldsymbol{x}_0, t_0))\,\mathrm{d}t_0, \tag{3.10}$$

corresponding to (3.5) and (3.7). Both of these expressions will have a use in our subsequent discussion.

Since the Reynolds number does not enter the analysis to a leading-order approximation, the net rate of transport in an oscillatory flow with frequency ω depends upon the values of (*a*) a properly defined Péclet number, and (*b*) the frequency parameter $\lambda = \omega a/U$. This is consistent with the well-known result that the rate of transport in a steady flow, which is recovered in the limit as λ tends to vanish, is independent of the Reynolds number to leading order in Pe.

In the following two sections, we shall compute the periodic function $\Phi(t)$ and its mean value over one period, for uniform oscillatory flow and for a family of oscillatory flows with linear variation in space, and shall discuss their dependence upon the frequency of the oscillation. In the limit of small frequencies, we shall recover the results for steady flow derived by previous authors.

4. Transport in uniform flow

We proceed to derive specific results for a particle that translates in a quiescent or a uniformly translating medium. In a frame of reference in which the particle appears to be stationary, the velocity of the fluid $\boldsymbol{U}(t)$ is assumed to be uniform but not necessarily constant in time.

4.1. *Green's functions of steady flow*

The Green's functions for steady flow are well known. Assuming that the velocity of translation $\boldsymbol{U}$ is time-independent, and using simplified forms of the general expressions for unsteady uniform or linear flows given in the Appendix, we obtain

$$G^{CD}(\boldsymbol{x}, \hat{t}; \boldsymbol{x}_0) = \frac{1}{(4\pi D\hat{t})^{3/2}} \exp\left(-\frac{|\boldsymbol{x}-\boldsymbol{x}_0-\boldsymbol{U}\hat{t}|^2}{4D\hat{t}}\right), \tag{4.1}$$

where $\hat{t} = t - t_0$. Integrating (4.1) with respect to t_0 from $-\infty$ up to the current time t, we derive the *stationary* Green's function with pole at $\boldsymbol{x}_0$:

$$\begin{aligned} G^{CD\text{-}S}(\boldsymbol{x}; \boldsymbol{x}_0) &= \frac{1}{(4\pi D)^{3/2}} \int_0^\infty \frac{1}{\hat{t}^{3/2}} \exp\left(-\frac{|\boldsymbol{x}-\boldsymbol{x}_0-\boldsymbol{U}\hat{t}|^2}{4D\hat{t}}\right) \mathrm{d}\hat{t} \\ &= \frac{1}{4\pi D|\boldsymbol{x}-\boldsymbol{x}_0|} \exp\left(-\frac{|\boldsymbol{x}-\boldsymbol{x}_0|\,|\boldsymbol{U}|}{2D}(1-\cos\theta)\right), \end{aligned} \tag{4.2}$$

where θ is the angle subtended between the vectors $\boldsymbol{x}-\boldsymbol{x}_0$ and $\boldsymbol{U}$. Expanding the exponential term on the right-hand side in a Taylor series for small values of its argument, we obtain the asymptotic expansion

$$G^{CD\text{-}S}(\boldsymbol{x}; \boldsymbol{x}_0) \approx \frac{1}{4\pi D|\boldsymbol{x}-\boldsymbol{x}_0|} - \frac{|\boldsymbol{U}|}{8\pi D^2}(1-\cos\theta) + \dots . \tag{4.3}$$

As the observation point $\boldsymbol{x}$ tends to the pole of the singularity $\boldsymbol{x}_0$, the terms represented by the dots tend to vanish.

4.2. *Late stages of transport in steady flow*

Substituting (4.3) into (2.11), we obtain the well-known result

$$F(\infty) = \tfrac{1}{2}Pe, \tag{4.4}$$

where $Pe = a|\boldsymbol{U}|/D$. Substituting (4.1) into (2.9) and (2.10), we obtain the less-known result

$$\frac{Q(t)-Q_0}{Q_0} = Nu_0\, Pe\frac{1}{2}\left(1+\frac{1}{2\pi^{1/2}}\int_{\xi}^{\infty}\frac{\mathrm{e}^{-\eta}}{\eta^{3/2}}\mathrm{d}\eta\right), \tag{4.5}$$

where $\xi = |\boldsymbol{U}|^2(t-t_s)/4D$. Evaluating the integral yields

$$\frac{Q(t)-Q_0}{Q_0} = Nu_0\, Pe\frac{1}{2}\left(1+\frac{\mathrm{e}^{-\xi}}{(\pi\xi)^{1/2}}-\operatorname{erfc}(\xi^{1/2})\right), \tag{4.6}$$

which is consistent with equation (32) of Choudhury & Drake (1971) for a spherical particle, derived by the method of the Laplace transform. This agreement serves to confirm the consistency of the present approach and the accuracy of the new results derived in §5 for a family of linear flows.

We have thus found that the rate of transport approaches its steady value at long times in an exponential manner. In the absence of fluid motion, $|\boldsymbol{U}| = 0$, we obtain the well-known result

$$\frac{Q(t)-Q_0}{Q_0} = Nu_0\frac{a}{(\pi D(t-t_s))^{1/2}} \tag{4.7}$$

corresponding to the limit of pure condition or diffusion. Contrasting the exponential decay shown in (4.6) with the inverse-square-root decay shown in (4.7) demonstrates the significant influence of the fluid motion.

4.3. *Uniform oscillatory flow*

We proceed to consider transport oscillatory flow with angular frequency ω, with the velocity field given by $\boldsymbol{U}(t) = \boldsymbol{W}+\boldsymbol{V}\sin(\omega t+\phi)$, where $\boldsymbol{W}$ is the mean value, $\boldsymbol{V}$ is the constant amplitude, and ϕ is the phase shift. The corresponding Green's function is readily found from (A 9) of the Appendix to be

$$G^{CD}(\boldsymbol{x},\hat{t};\boldsymbol{x}_0) = \frac{1}{(4\pi D\hat{t})^{3/2}} \times\exp\left(-\frac{1}{4D\hat{t}}\left|\boldsymbol{x}-\boldsymbol{x}_0-\boldsymbol{W}\hat{t}+\boldsymbol{V}\frac{\cos(\omega t+\phi)-\cos(\omega t_0+\phi)}{\omega}\right|^2\right), \tag{4.8}$$

where $\hat{t} = t-t_0$. The quantity $\Delta T(t)$ computed from (3.7) is given by

$$\Delta T = \alpha Q_0\, C(t)+\alpha Q_0\frac{\omega^{1/2}}{(4\pi D)^{3/2}}\int_{-\infty}^{\omega t}\left\{\exp\left(-\frac{\hat{t}}{4D}|\boldsymbol{W}+\boldsymbol{V}\sin(\omega t+\phi)|^2\right) -\exp\left(-\frac{\hat{t}}{4D}\left|\boldsymbol{W}-\boldsymbol{V}\frac{\cos(\omega t+\phi)-\cos(\omega t_0+\phi)}{\omega\hat{t}}\right|^2\right)\right\}\frac{\mathrm{d}\omega t_0}{(\omega\hat{t})^{3/2}}. \tag{4.9}$$

Using (4.3), we find

$$C(t) = \frac{1-\cos\theta}{8\pi D^2}|\boldsymbol{W}+\boldsymbol{V}\sin(\omega t+\phi)|. \tag{4.10}$$

The function $\Phi(t)$ then follows from the right-hand side of (3.10) as

$$\Phi(t) = \frac{a}{2D}|\boldsymbol{W}+\boldsymbol{V}\sin(\omega t+\phi)|+\frac{a\omega^{1/2}}{(4\pi D)^{1/2}}\int_{-\infty}^{\omega t}\left\{\exp\left(-\frac{\hat{t}}{4D}|\boldsymbol{W}+\boldsymbol{V}\sin(\omega t+\phi)|^2\right) -\exp\left(-\frac{\hat{t}}{4D}\left|\boldsymbol{W}-\boldsymbol{V}\frac{\cos(\omega t+\phi)-\cos(\omega t_0+\phi)}{\omega\hat{t}}\right|^2\right)\right\}\frac{\mathrm{d}\omega t_0}{(\omega\hat{t})^{3/2}}. \tag{4.11}$$

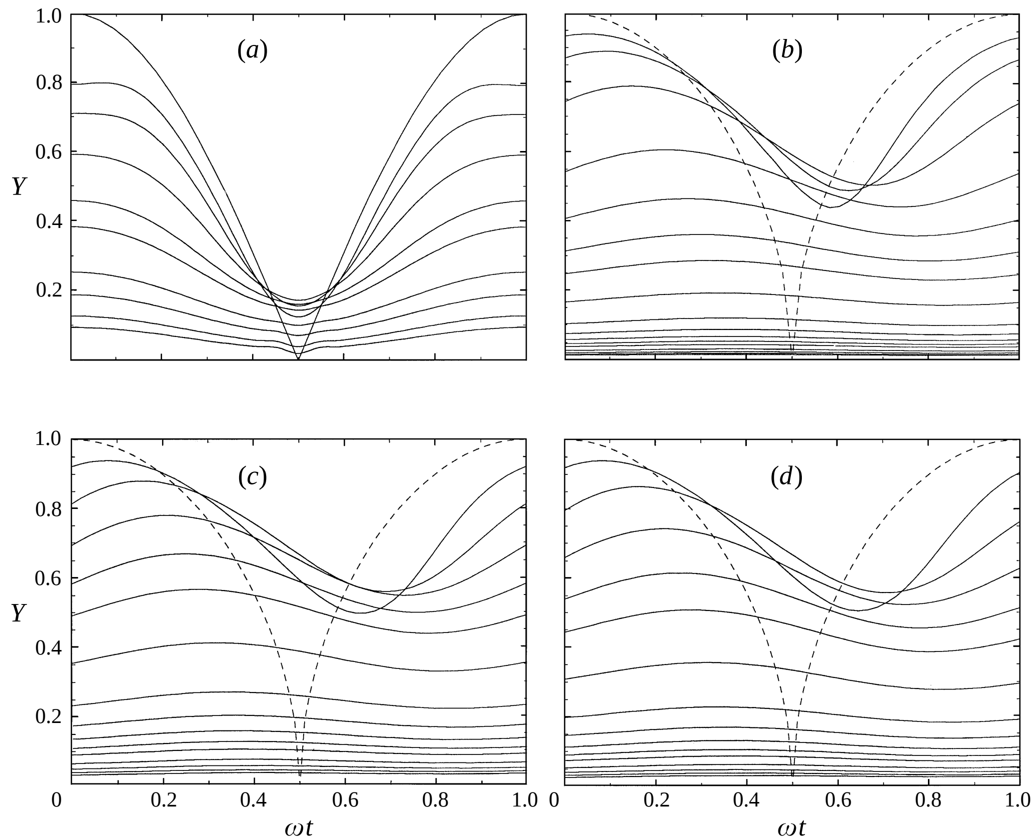

FIGURE 3. Transport in an oscillatory flow. Graphs of the periodic function $Y(\omega t, \delta)$ whose mean value expresses the enhancement of the rate of scalar transport, for several values of the frequency parameter δ, and phase shift $\phi = \pi/2$. The dashed lines represent the limit of quasi-steady transport corresponding to $\delta = 0$, expressed by the function $|\cos(\omega t)|$. (*a*) Uniform oscillatory flow with $\delta =$ 0.05, 0.10, 0.20, 0.40, 0.60, 1.4, 2.5, 5.0, 8.0; (*b*) simple shear flow with $\delta =$ 0.05, 0.10, 0.20, 0.40, 0.60, 0.80, 1.0, 1.4, 2.0, 2.5, 3.0, 3.5, 4.0, 5.0, 6.0, 7.0, 8.0; (*c*) two-dimensional extensional flow with $\delta =$ 0.20, 0.40, 0.60, 0.80, 1.0, 1.4, 2.0, 2.5, 3.0, 3.5, 4.0, 5.0, 6.0, 7.0, 8.0; (*d*) axisymmetric extensional flow for the same values of δ as in (*b*).

The first term on the right-hand side describes the limit of quasi-steady transport.

Let us assume, for simplicity, that the mean velocity vanishes, that is, $|\boldsymbol{W}| = 0$. We introduce the Péclet number $Pe = a|\boldsymbol{V}|/D$, and define the frequency parameter $\delta = \lambda/Pe = \omega D/|\boldsymbol{V}|^2$ where $\lambda = a\omega/|\boldsymbol{V}|$ is a purely hydrodynamic frequency parameter. In this case (4.10) can be placed in the form

$$\Phi(t) = \tfrac{1}{2} Pe\, Y(\omega t, \delta), \tag{4.12}$$

where

$$Y(\omega t, \delta) = |\sin(\omega t + \phi)| + \frac{\delta^{1/2}}{\pi^{1/2}} \int_{-\infty}^{\omega t} \left\{ \exp\left[-\frac{\omega \hat{t}}{4\delta} \sin^2(\omega t + \phi) \right] - \exp\left(-\frac{\omega \hat{t}}{4\delta} \left[\frac{\cos(\omega t + \phi) - \cos(\omega t_0 + \phi)}{\omega \hat{t}} \right]^2 \right) \right\} \frac{\mathrm{d}\omega t_0}{(\omega \hat{t})^{3/2}}, \tag{4.13}$$

and $\hat{t} = t - t_0$.

In figure 3(*a*), we plot the dimensionless function $Y(\omega t, \delta)$ with respect to ωt over one

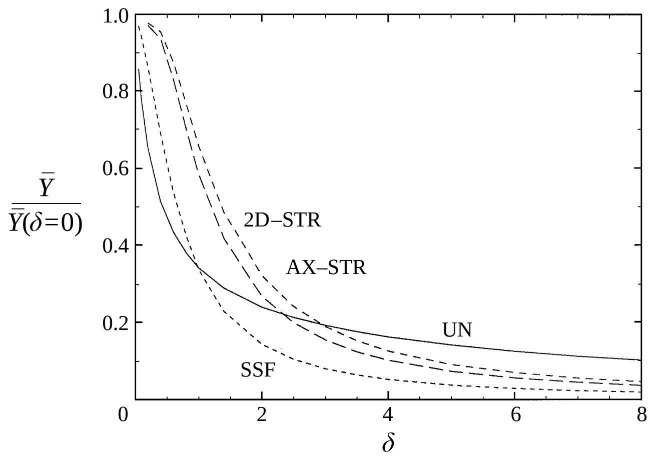

FIGURE 4. The mean value of the function $Y(\omega t, \delta)$ over one period, expressing the effective enhancement in the rate of scalar transport due to an oscillatory flow, as a function of the frequency parameter δ defined in table 1. Solid line is for uniform oscillatory flow, short-dashed line for simple shear flow, longer-dashed line for two-dimensional extensional flow, and the longest-dashed line is for axisymmetric extensional flow.

period for several values of the dimensionless frequency δ, and for $\phi = \pi/2$ corresponding to $\boldsymbol{U}(t) = \boldsymbol{V}\cos(\omega t)$. As δ tends to vanish, $Y(\omega t, \delta)$ behaves like $|\cos(\omega t)|$, plotted with the dashed line, corresponding to quasi-steady transport. As δ is increased, the range of variation of Y is reduced significantly: the oscillatory motion effectively smooths out the gradients of the transported scalar over one period, and enhances the transport rate even when the instantaneous shear rate passes through its null point.

In figure 4, we plot with a solid line the mean value of $Y(\omega t, \delta)$ over one period, as a function of δ, normalized by the quasi-steady value corresponding to $\delta = 0$, and obtain a monotonically decaying curve, which demonstrates that the oscillatory motion has a strong influence on the effective rate of transport. A flow with $\delta = 0.50$ reduces the mean rate of transport by a factor of nearly 2. In physical terms, the oscillatory flow homogenizes the scalar field and thereby reduces the magnitude of its effective gradient owing to mixing.

Unfortunately, a comparison between the predictions of these results with experimental observations, such as those by Gibert & Angelino (1973), has not been possible. The experiments are typically conducted at high Péclet numbers, and the values of the physical parameters are not well documented.

5. Transport in linear flows

We turn next to considering transport from a neutrally buoyant particle that is convected with the fluid. In a frame of reference in which the particle appears to be stationary, the ambient velocity field is given by $\boldsymbol{u}^{\infty}(t) = \boldsymbol{A}(t)\cdot(\boldsymbol{x}-\boldsymbol{x}_0)$, where $\boldsymbol{x}_0$ is the designated particle centre.

5.1. *Steady transport in steady flows*

To establish a point of reference for the forthcoming results, we briefly discuss certain well-known results for steady transport in a steady flow. Leaving aside the case of purely rotational flow – which has no effect on the rate of transport to leading order in the Péclet number for an arbitrary particle, and to all orders in the Péclet number for a spherical particle – we adopt a scaling introduced by Batchelor (1979), and express the asymptotic value of the dimensionless function $F(t)$ at large times, defined in (2.11) as

$$F(\infty) = a\frac{(\boldsymbol{E}:\boldsymbol{E})^{1/4}}{(4\pi D)^{1/2}}\chi, \tag{5.1}$$

where $\boldsymbol{E}$ is the rate-of-deformation tensor, and χ is a dimensionless constant. This equation is essentially a definition for χ. Using the expression for the Green's function given in (A 9) of the Appendix, we find that, for an incompressible fluid,

$$\chi = \frac{1}{(\boldsymbol{E}:\boldsymbol{E})^{1/4}}\int_{-\infty}^{t}\left(\frac{1}{\hat{t}^{3/2}} - \frac{D^{3/2}}{\beta^{1/2}}\right)\mathrm{d}t_0, \tag{5.2}$$

where $\hat{t} = t - t_0$ and β is defined in (A 11). An approximate value of χ can be obtained on the basis of (A 19). The result is

$$\chi \approx \frac{1}{12}\int_0^\infty \frac{p^{1/2}}{1+\frac{1}{12}p^2}\mathrm{d}p = 3^{-1/4}\frac{\pi}{2} = 1.194, \tag{5.3}$$

accurate to the digits shown, independently of the nature of the flow, as long as it is not a purely rotational flow (Gradshteyn & Ryzhik 1980, p. 369; Batchelor 1979, p. 381).

5.2. *Late stages of transport in steady flow*

To derive the functional form of the rate of transport from a particle that is suddenly immersed into the fluid near the steady state, we combine (2.10) and (2.11) with the expression for the Green's function gives in (A 9) and find

$$\frac{Q(t)-Q_0}{Q_0} = Nu_0\left(F(\infty) + \frac{aD}{(4\pi)^{1/2}}\int_{t-t_s}^{\infty}\frac{\mathrm{d}\hat{t}}{\beta^{1/2}}\right). \tag{5.4}$$

5.3. *Mean rate of transport in oscillatory flow*

The dimensionless function $\Phi(t)$, evaluated from (3.9), is given by

$$\Phi(t) = \frac{a}{(4\pi D)^{1/2}}\int_{-\infty}^{t}\left(\frac{1}{\hat{t}^{3/2}} - \frac{D^{3/2}}{\beta^{1/2}}\right)\mathrm{d}t_0. \tag{5.5}$$

In the following subsections, we shall evaluate the integrals on the right-hand sides of (5.4) and (5.5) for simple shear flow and purely straining two-dimensional or axisymmetric flow.

5.4. *Transport in steady simple shear flow*

Consider steady simple shear flow along the x-axis, varying along the y-axis. The unperturbed incident velocity field is given by $\boldsymbol{u}^\infty = (\gamma(y-y_0), 0, 0)$, where γ is a constant shear rate. Solving equation (A 4) of the Appendix for $\boldsymbol{B}$, we find that it is equal to the identity matrix except that $B_{xy} = -\gamma\hat{t}$. Using (A 7) and (A 10) we then obtain

$$\boldsymbol{J} = D\hat{t}\begin{bmatrix} 1+\frac{1}{3}\gamma^2\hat{t}^2 & -\frac{1}{2}\gamma\hat{t} & 0 \\ -\frac{1}{2}\gamma\hat{t} & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \tag{5.6}$$

from which it follows that

$$\beta = D^3\hat{t}^3(1+\tfrac{1}{12}\gamma^2\hat{t}^2). \tag{5.7}$$

Equation (5.2) with $\boldsymbol{E}:\boldsymbol{E} = \gamma^2/2$ then yields

$$\chi = 2^{1/4}\int_0^\infty \left(1-\frac{1}{(1+\frac{1}{12}p^2)^{1/2}}\right)\frac{\mathrm{d}p}{p^{3/2}} = 1.083, \tag{5.8}$$

accurate to the digits shown, in agreement with the results of Frankel & Acrivos (1968). It is interesting to note that this value is lower than the general approximate value predicted from (5.3) by only 9%. Equation (5.1) takes the specific form

$$F(\infty) = 0.257\, Pe^{1/2}, \tag{5.9}$$

where $Pe = \gamma a^2/D$.

Substituting the expressions given in (5.7) and (5.9) into (5.4), and evaluating the asymptotic form of the integral at large times, we find

$$\frac{Q(t)-Q_0}{Q_0} = Nu_0\, Pe^{1/2}\left(0.257+\frac{2}{(3\pi)^{1/2}\gamma^{3/2}(t-t_s)^{3/2}}+\dots\right), \tag{5.10}$$

where the dots represent terms with faster decay. The algebraic inverse 3/2 decay of the transient component may be contrasted with the exponential decay corresponding to uniform flow shown in (4.6). The physical implication of this difference will be discussed in the concluding section.

5.5. *Oscillatory simple shear flow*

We assume now that the shear rate oscillates harmonically in time with angular frequency ω, so that $u_x(t) = \bar{\gamma}\sin(\omega t+\phi)(y-y_0)$, $u_y = 0$, $u_z = 0$, where $\bar{\gamma}$ is a constant amplitude, and ϕ is the phase shift. This flow occurs, for example, when a small neutrally buoyant particle is suspended in a fluid between two parallel plates, where one or both of the plates oscillate parallel to themselves in a periodic fashion (Leighton 1989).

Integrating (A 4) we find that $\boldsymbol{B}(t)$ is equal to the identity matrix except that

$$B_{xy}(t) = (\cos(\omega t+\phi)-\cos(\omega t_0+\phi))/\delta, \tag{5.11}$$

where $\delta = \omega/\bar{\gamma}$ is a dimensionless frequency parameter. The modified diffusivity is readily found from (A 7) to be

$$\boldsymbol{S}(t) = D\begin{bmatrix} 1+\dfrac{[\cos(\omega t+\phi)-\cos(\omega t_0+\phi)]^2}{\delta^2} & \dfrac{\cos(\omega t+\phi)-\cos(\omega t_0+\phi)}{\delta} & 0 \\ \dfrac{\cos(\omega t+\phi)-\cos(\omega t_0+\phi)}{\delta} & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{5.12}$$

Straightforward integration as dictated by (A 10) gives

$$\left.\begin{aligned} J_{xx} &= D\hat{t}+\frac{D}{\omega\delta^2}\{\tfrac{1}{2}\omega\hat{t}+\tfrac{1}{4}[\sin(2\omega t+2\phi)-\sin(2\omega t_0+2\phi))] \\ &\quad -2\cos(\omega t_0+\phi)[\sin(\omega t+\phi)-\sin(\omega t_0+\phi)]+2\omega\hat{t}\cos^2(\omega t_0+\phi)]\}, \\ J_{xy} &= J_{xy} = \frac{D}{\omega\delta}[\sin(\omega t+\phi)-\sin(\omega t_0+\phi)-\omega\hat{t}\cos(\omega t_0+\phi)], \\ J_{yy} &= J_{zz} = D\hat{t}, \quad J_{xz} = J_{zx} = J_{yz} = J_{zy} = 0. \end{aligned}\right\} \tag{5.13}$$

In the limit as t tends to t_0, these expressions reduce to those shown in (5.6) with $\gamma = \bar{\gamma} \sin(\omega t_0 + \phi)$.

It now follows readily that

$$\beta \equiv \mathrm{Det}(\boldsymbol{J}) = D^3 \hat{t}^3 \zeta(\omega t, \omega t_0, \delta), \tag{5.14}$$

where the dimensionless function ζ is given by

$$\zeta(\omega t, \omega t_0, \delta) = 1 + \frac{1}{\delta^2}\frac{1}{2} \left[1 + \frac{\sin 2(\omega t + \phi) - \sin 2(\omega t_0 + \phi)}{2\omega(t - t_0)} - 2 \left(\frac{\sin(\omega t + \phi) - \sin(\omega t_0 + \phi)}{\omega(t - t_0)} \right)^2 \right]. \tag{5.15}$$

In the limit as t tends to t_0, we obtain the asymptotic expression

$$\zeta \approx 1 + \tfrac{1}{12} \sin^2(\omega t + \phi)\, \bar{\gamma}^2 \hat{t}^2 + \tfrac{5}{24} \delta \sin[2(\omega t + \phi)]\, \bar{\gamma}^3 \hat{t}^3. \tag{5.16}$$

To explore the asymptotic behaviour at low frequencies we take the limit of the right-hand side of (5.15) as δ tends to vanish, while $\bar{\gamma} t$ and $\bar{\gamma} t_0$ are held constant, and recover (5.16).

Substituting (5.14) into (5.5), we find that the dimensionless function $\Phi(t)$ is given by

$$\Phi(t) = \frac{Pe^{1/2}}{2^{1/4}(4\pi)^{1/2}}\, \chi\, Y(\omega t, \delta), \tag{5.17}$$

where $Pe = \bar{\gamma} a^2 / D$. The value of the dimensionless constant χ is given in (5.8), and the dimensionless function Y is defined as

$$Y(\omega t, \delta) = \frac{2^{1/4}}{\chi} \delta^{1/2} \int_0^\infty \left(1 - \frac{1}{[\zeta(\omega t, \omega t - p, \delta)]^{1/2}} \right) \frac{\mathrm{d}p}{p^{3/2}}. \tag{5.18}$$

In figure 3(*b*), we plot $Y(\omega t, \delta)$ with respect to ωt over one period for several values of the dimensionless frequency δ, for $\phi = \pi/2$ corresponding to $\gamma = \bar{\gamma} \cos(\omega t)$. As δ tends to vanish, $Y(\omega t, \delta)$ behaves like $|\cos(\omega t)|^{1/2}$, which is represented by the dashed line, corresponding to the limit of quasi-steady transport. As δ is increased, the range of variation of Y is reduced, which shows that the oscillatory motion effectively smooths out the gradients of the scalar over each period, and enhances the transport rate even when the instantaneous shear rate passes through its null point. Furthermore, as δ is increased, an increasingly larger phase shift is established between the shear rate and the function $Y(\omega t, \delta)$. But since $Y(\omega t, \delta)$ is not proportional to the local rate of transport, the significance of these behaviours is unclear.

In figure 4, we plot with a short dashed line the mean value of $Y(\omega t, \delta)$ over one period as a function of δ, normalized by its value at steady state corresponding to $\delta = 0$, and obtain a monotonically decaying curve. The general features of the graph and the physical interpretation of its behaviour are similar to those discussed in the preceding section for uniform flow.

5.6. *Purely straining flows*

Next, we consider transport in a purely straining flow. The velocity gradient tensor $\boldsymbol{A}$ is symmetric with vanishing trace, and the rate-of-deformation tensor $\boldsymbol{E}$ is equal to $\boldsymbol{A}$.

Considering first transport in a steady flow, we use (A 7) and (A 10) and find

$$\boldsymbol{J}(\hat{t}) = \tfrac{1}{2} D \boldsymbol{E}^{-1} \cdot [\boldsymbol{I} - \exp(-2\boldsymbol{E}\hat{t})] \tag{5.19}$$

from which it follows that

$$\beta = \frac{D^3}{8}\frac{\mathrm{Det}\,[\boldsymbol{I} - \exp(-2\boldsymbol{E}\hat{t})]}{\mathrm{Det}\,(\boldsymbol{E})}. \tag{5.20}$$

It is convenient to refer to the principal axes of $\boldsymbol{E}$ in which the tensor $\boldsymbol{J}$ is diagonal with elements given by

$$J_{ii}(\hat{t}) = D\hat{t}\,\frac{1 - \exp(-2E_{ii}\,\hat{t})}{2E_{ii}\,\hat{t}}, \tag{5.21}$$

where summation is not implied over i. The determinant of $\boldsymbol{J}$ follows readily as

$$\beta \equiv \mathrm{Det}\,(\boldsymbol{J}) = D^3\hat{t}^3\,\frac{\sinh(E_{11}\,\hat{t})\sinh(E_{22}\,\hat{t})\sinh(E_{33}\,\hat{t})}{(E_{11}\,\hat{t})(E_{22}\,\hat{t})(E_{33}\,\hat{t})}, \tag{5.22}$$

which is consistent with the asymptotic expansion for short times shown in (A 19).

Two-dimensional extensional flow corresponds to $E_{11} = \gamma$, $E_{22} = -\gamma$, $E_{33} = 0$, where γ is the strain rate. Taking the limit of (5.22) as E_{33} tends to vanish, we find

$$\beta = D^3\hat{t}\,\frac{\cosh(2\gamma\hat{t}) - 1}{2\gamma^2}. \tag{5.23}$$

Axisymmetric extension flow corresponds to $E_{11} = \gamma$, $E_{22} = \gamma$, $E_{33} = -2\gamma$. Equation (5.22) gives

$$\beta = \frac{D^3}{8\gamma^3}(\sinh(4\gamma\hat{t}) - 2\sinh(2\gamma\hat{t})). \tag{5.24}$$

Substituting the right-hand side of (5.22) into (5.2), we find

$$\chi = \frac{1}{(\boldsymbol{E}:\boldsymbol{E})^{1/4}}\int_0^\infty \left(1 - \left[\frac{E_{11}\,E_{22}\,E_{33}\,\hat{t}^3}{\sinh(E_{11}\,\hat{t})\sinh(E_{22}\,\hat{t})\sinh(E_{33}\,\hat{t})}\right]^{1/2}\right)\frac{\mathrm{d}\hat{t}}{\hat{t}^{3/2}}. \tag{5.25}$$

In both the two-dimensional and axisymmetric extensional flow cases, the numerical evaluation of the integral in (5.25) gives $\chi = 1.28$ (Batchelor 1979), which is predicted with good accuracy by the approximate form shown in (5.3). Using (5.1) we then obtain

$$F(\infty) = 1.36Pe^{1/2}, \quad F(\infty) = 2.00\,Pe^{1/2} \tag{5.26}$$

respectively for two-dimensional and axisymmetric flow, where $Pe = \gamma a^2/D$. Note that the values of the coefficients on the right-hand sides are significantly higher than that for simple shear flow shown in (5.9).

5.7. *Late stages of transport in steady purely straining flows*

Substituting the expressions given in (5.23), (5.24), and (5.26) into (5.4), and evaluating the integral at large times we find that, for two-dimensional purely straining flow,

$$\frac{Q(t) - Q_0}{Q_0} = Nu_0\,Pe^{1/2}(1.36 + \mathrm{erfc}\,(\gamma(t - t_s)^{1/2}) + \ldots) \tag{5.27}$$

and for axisymmetric purely straining flow,

$$\frac{Q(t) - Q_0}{Q_0} = Nu_0\,Pe^{1/2}\left(2.00 + \frac{1}{\pi^{1/2}}\exp(-2\gamma(t - t_s)) + \ldots\right). \tag{5.28}$$

In both cases, we obtain an exponential approach to the steady state.

5.8. *Transport in oscillatory purely straining flows*

In the final case study, we consider oscillatory straining flow with the time-dependent velocity gradient tensor $\boldsymbol{A}(t) = \bar{\gamma}\boldsymbol{M}\sin(\omega t+\phi)$, where $\bar{\gamma}$ is the amplitude of the rate of strain, $\boldsymbol{M}$ is a symmetric dimensionless matrix with vanishing trace, ω is the angular frequency of the oscillations, and ϕ is the phase shift.

Two-dimensional extensional flow corresponds to $M_{11} = 1$, $M_{22} = -1$, $M_{33} = 0$, and axisymmetric extensional flow corresponds to $M_{11} = M_{22} = 1$, $M_{33} = -2$. The Green's function of the associated convection–diffusion equation in these two cases was derived by Krishnan & Leighton (1992) using a different method. Using the general solution given in (A 13), we find

$$\boldsymbol{B}(t) = \exp\{\boldsymbol{M}[\cos(\omega t+\phi) - \cos(\omega t_0+\phi)]/\delta\}, \tag{5.29}$$

where $\delta = \omega/\bar{\gamma}$ is a dimensionless frequency parameter. The modified diffusivity follows from (A 7) as

$$\boldsymbol{S}(t) = D\exp\{\boldsymbol{M}2[\cos(\omega t+\phi) - \cos(\omega t_0+\phi)]/\delta\} \tag{5.30}$$

and this can be integrated according to (A 10) to give

$$\boldsymbol{J}(t) = D\hat{t}\,\boldsymbol{H}(\omega t, \omega t_0, \delta), \tag{5.31}$$

where the dimensionless function $\boldsymbol{H}$ is defined as

$$\boldsymbol{H}(\omega t, \omega t_0\,\delta) = \frac{1}{\omega(t-t_0)}\int_{\omega t_0}^{\omega t} \exp\{\boldsymbol{M}2[\cos(\omega t+\phi) - \cos(\omega t_0+\phi)]/\delta\}\,\mathrm{d}\omega t_0. \tag{5.32}$$

The determinant of $\boldsymbol{J}$ may be expressed in the form

$$\beta = D^3\hat{t}^3\zeta(\omega t, \omega t_0, \delta) \tag{5.33}$$

where $\zeta(\omega t, \omega t_0, \delta) = \mathrm{Det}[\boldsymbol{H}(\omega t, \omega t_0, \delta)]$ is a dimensionless function. As ωt_0 tends to ωt, ζ approaches the value of unity independently of the value of δ.

When the Cartesian axes point in the principal direction of $\boldsymbol{E}$, the tensor $\boldsymbol{H}$ is diagonal. In the limit as the dimensionless frequency δ tends to vanish, the diagonal elements of $\boldsymbol{H}$ tend to the fractions given on the right-hand side of (5.21), and the function ζ tends to the fraction given on the right-hand side of (5.22) with $\boldsymbol{E} = \bar{\gamma}\boldsymbol{M}\sin(\omega t_0+\phi)$.

Substituting (5.33) into (5.5), we find

$$\Phi(t) = \left(\frac{Pe}{4\pi}\right)^{1/2}(\boldsymbol{M}\mathbin{:}\boldsymbol{M})^{1/4}\chi\, Y(\omega t, \delta), \tag{5.34}$$

where $Pe = \bar{\gamma}a^2/D$, and χ is given by (5.25) with the matrix $\bar{\gamma}\boldsymbol{M}$ in place of $\boldsymbol{E}$. The dimensionless function Y is defined as

$$Y(\omega t, \delta) = \frac{\delta^{1/2}}{\chi(\boldsymbol{M}\mathbin{:}\boldsymbol{M})^{1/4}}\int_0^\infty \left(1 - \frac{1}{[\zeta(\delta, \omega t, \omega t - p)]^{1/2}}\right)\frac{\mathrm{d}p}{p^{3/2}}. \tag{5.35}$$

In figure 3 (c, d), we plot the function $Y(\omega t, \delta)$ with respect to ωt over one period for $\phi = \pi/2$ corresponding to $\boldsymbol{A}(t) = \bar{\gamma}\,\boldsymbol{M}\cos(\omega t)$, for several values of the dimensionless frequency δ, and for two-dimensional and axisymmetric elongational flow. In both cases, as δ tends to vanish, $Y(\omega t, \delta)$ behaves like $|\cos(\omega t)|^{1/2}$, which is shown with a dashed line, representing the limit of quasi-steady transport. In figure 4, we plot with a dashed line and with a long-dashed line the corresponding mean values of $Y(\omega t, \delta)$ over one period. The general features of the graphs and their physical interpretation are similar to those discussed previously for simple shear flow, although quantitative differences are apparent.

6. Discussion

We used the method of matched asymptotic expansions to study the effect of a steady or oscillatory flow on the unsteady rate of scalar transport from a suspended particle, to leading order with respect to the Péclet number. A summary of definitions and prior and new results is presented in table 1. Note that, in all cases, the enhancement in the rate of transport is proportional to the effective particle size a.

For a particle that has been released into a steady flow, we found that the rate of transport approaches its asymptotic value at long times in a manner that depends upon the structure of the incident flow. For uniform and purely elongation flow we found an exponential approach, whereas for simple shear flow consisting of an elongational and a rotational component we found an algebraic approach with an exponent of $-3/2$. These differences suggest that the rate of decay in a general incident flow with a non-zero rotational component will be algebraic with an exponent of $-3/2$. A purely rotational flow causes the fluid parcels to move over concentric spherical surfaces centred at the instantaneous particle centre. The corresponding rate of transport is expected to decay in an algebraic manner with the diffusive exponent of $-1/2$.

An oscillatory flow homogenizes the scalar field and thus decelerates the rate of transport with respect to that, that would prevail in a corresponding steady flow, by a factor that depends on a properly defined frequency parameter δ, as shown in figure 4. The definition of δ for each type of flow is presented in table 1. The results reveal that a simple shear flow has a stronger effect on the overall rate of transport than a purely straining two-dimensional or axisymmetric flow, but this ranking depends on the particular way of defining the shear rate. To leading order in the Péclet number, a purely rotational oscillatory flow has no influence on either the instantaneous or the mean rate of transport.

The theory developed provides us with an expression for the enhancement in the rate of transport with respect to its purely diffusive value, to leading order with respect to the Péclet number, in terms of the purely diffusive value. This is done by considering the behaviour of a point source close to its pole, and identifying its effect on the boundary condition that is imposed on the scalar distribution at the end of the inner regime where convective effects start making a significant contribution. Similar analyses allow us to compute the Oseen force exerted on a translating particle in terms of the fundamental velocity field associated with an Oseenlet, that is, a point force (Brenner 1961), and the force exerted on a vibrating particle in linearized flow in terms of the velocity field due to an unsteady Stokeslet, that is, an oscillatory point force (Pozrikidis 1989). The results are expressed in terms of the resistance matrices for Stokes flow, just as the present results are expressed in terms of the purely diffusive rate of transport.

The problem of unsteady flow past a suspended particle has many similarities with the problem of oscillatory shear flow over a hot film on a probe that is mounted flush on an insulated solid boundary (Pedley 1972, 1976). If the flow occurs at high Reynolds numbers, and if the transport occurs at large Péclet numbers, measurements of the rate

Velocity	Steady flow	Oscillatory flow
	$\dfrac{Q-Q_0}{Q_0} = Nu_0\,F(t)$	$\dfrac{Q-Q_0}{Q_0} = Nu_0\,\bar{\Phi}$
Uniform flow		
$u_x = U$	$F(\infty) = \frac{1}{2}Pe$	$U = V\sin(\omega t+\phi)$
$u_y = 0$	$Pe = Ua/D$	$Pe = Va/D$
$u_z = 0$	See also (4.6)	$\delta = \omega D/V^2$
		See (4.12)
		and figures 3 and 4
Simple shear flow		
$u_x = \gamma(y-y_0)$	$F(\infty) = 0.257\,Pe^{1/2}$	$\gamma = \bar{\gamma}\sin(\omega t+\phi)$
$u_y = 0$	$Pe = \gamma a^2/D$	$Pe = \bar{\gamma}a^2/D$
$u_z = 0$	See also (5.10)	$\delta = \omega/\bar{\gamma}$
		See (5.17)
		and figures 3 and 4
Two-dimensional straining flow		
$u_x = \gamma(x-x_0)$	$F(\infty) = 1.36\,Pe^{1/2}$	$\gamma = \bar{\gamma}\sin(\omega t+\phi)$
$u_y = -\gamma(y-y_0)$	$Pe = \gamma a^2/D$	$Pe = \bar{\gamma}a^2/D$
$u_z = 0$	See also (5.28)	$\delta = \omega/\gamma$
		See (5.34)
		and figures 3 and 4
Axisymmetric straining flow		
$u_x = \gamma(x-x_0)$	$F(\infty) = 2.00\,Pe^{1/2}$	$\gamma = \bar{\gamma}\sin(\omega t+\phi)$
$u_y = \gamma(y-y_0)$	$Pe = \gamma a^2/D$	$Pe = \bar{\gamma}a^2/D$
$u_z = -2\gamma(z-z_0)$	See also (5.28)	$\delta = \delta/\bar{\gamma}$
		See (5.34)
		and figures 3 and 4

TABLE 1. A summary of definitions and results.

of heat transfer can be used to deduce the magnitude of the wall shear stress. Pedley (1972, 1976) developed a theory for computing the instantaneous rate of transport in a general unsteady and oscillatory simple shear flow, and Kaiping (1983) carried out numerical computations. The results of Kaiping (1983) regarding the averaged heat flux in a reversing pulsating flow, shown in his figure 12(*b*), are qualitatively similar to those shown in figure 4 of this paper, even though the transport occurs under different conditions.

The present theory can be applied with straightforward changes in symbols and notation to study the aforementioned problem studied by Pedley in the limit of small Péclet numbers. The analysis will allow us to compute the late stages of transport from a film that is mounted flush with the surface of a wall, subject to a step increase in its temperature, as well as the mean rate of transport in oscillatory flow. Obtaining quantitative results, however, requires the availability of the Green's function for semi-infinite flow, whose derivation and computation are elusive.

The Department of Chemical Engineering of the Imperial College and, in particular, Professor Chris Lawrence, provided me with a hospitable environment for carrying out parts of this research. Financial support was provided by the National Science Foundation and the SUN Microsystems Corporation. Acknowledgment is made to the Donors of the Petroleum Research Fund, administered by the American Chemical Society, for partial support of this research.

Appendix. Green's function of the convection–diffusion equation for unsteady uniform or linear flow

In this Appendix, we derive the Green's function of the unsteady convection–diffusion equation for an unsteady linear flow with velocity $\boldsymbol{v}(t) = \boldsymbol{A}(t)\cdot\boldsymbol{x}+\boldsymbol{U}(t)$, representing the scalar field due to an impulsive point source. To broaden the applicability of the results, we allow the diffusivity to vary in time and have an anisotropic form expressed by the symmetric diffusivity tensor $\boldsymbol{D}(t)$, which is assumed to be independent of position in the domain of flow.

The Green's function satisfies the generalized convection–diffusion equation

$$\frac{\partial G^{CD}}{\partial t}+\nabla\cdot(\boldsymbol{v}(t)\,G^{CD}) = \boldsymbol{D}(t)\colon\nabla\nabla G^{CD}+\delta(x-y_0)\,\delta(y-y_0)\,\delta(z-z_0)\,\delta(t-t_0), \quad \text{(A 1)}$$

where δ is the one-dimensional delta function. As a first step towards computing the solution, we introduce Lagrangian coordinates, designated with a hat, that reduce (A 1) to an equivalent linear diffusion–reaction equation. The Lagrangian coordinates are related to the Eulerian coordinates by the transformation $\partial\hat{x}_i/\partial x_j = B_{ij}(\boldsymbol{x},t)$ where $\boldsymbol{B}$ is the inverse of the deformation gradient $\boldsymbol{F}$, $\boldsymbol{B}=\boldsymbol{F}^{-1}$. Since the deformation gradient associated with a linear flow is uniform, we write

$$\hat{\boldsymbol{x}} = \boldsymbol{B}(t)\cdot\boldsymbol{x}-\boldsymbol{c}(t), \quad \text{(A 2)}$$

where $\boldsymbol{c}(t)$ is a time-dependent vector to be determined as part of the solution. Using the chain rule of differentiation, we obtain the relations

$$\left.\begin{aligned}&\frac{\partial G^{CD}}{\partial t} = \frac{\partial G^{CD}}{\partial\hat{t}}+\left(\frac{\mathrm{d}\boldsymbol{B}}{\mathrm{d}t}\cdot\boldsymbol{x}-\frac{\mathrm{d}\boldsymbol{c}}{\mathrm{d}t}\right)\cdot\hat{\nabla}G^{CD},\\ &\nabla G^{CD} = \boldsymbol{B}^T(t)\cdot\hat{\nabla}G^{CD}, \quad \nabla\nabla G^{CD} = \boldsymbol{B}^T(t)\cdot(\hat{\nabla}\hat{\nabla}G^{CD})\cdot\boldsymbol{B}(t),\end{aligned}\right\} \quad \text{(A 3)}$$

where $\hat{t} = t-t_0$, and the hat over the gradient indicates differentiation with respect to the Lagrangian coordinates. It is understood that the partial derivative with respect to $\hat{t}$ is taken, keeping the Lagrangian spatial coordinates constant. Substituting (A 3) into (A 1), and requiring that $\boldsymbol{B}$ and $\boldsymbol{c}$ evolve according to the ordinary differential equations

$$\mathrm{d}\boldsymbol{B}/\mathrm{d}t+\boldsymbol{B}\cdot\boldsymbol{A} = \boldsymbol{0} \quad \text{(A 4)}$$

and

$$\mathrm{d}\boldsymbol{c}/\mathrm{d}t-\boldsymbol{B}\cdot\boldsymbol{U} = \boldsymbol{0}, \quad \text{(A 5)}$$

reduces (A 1) to a linear diffusion equation with a homogeneous linear sink term forced by the uniform rate of expansion,

$$\frac{\partial G^{CD}}{\partial\hat{t}}+G^{CD}\,\nabla\cdot\boldsymbol{v}(t) = \boldsymbol{S}(t)\colon\hat{\nabla}\hat{\nabla}G^{CD}+\delta(x-x_0)\,\delta(y-y_0)\,\delta(z-z_0)\,\delta(\hat{t}), \quad \text{(A 6)}$$

where $\boldsymbol{S}(t)$ is a time-dependent symmetric modified diffusivity tensor given by

$$\boldsymbol{S}(t) = \boldsymbol{B}(t)\cdot\boldsymbol{D}(t)\cdot\boldsymbol{B}^T(t). \quad \text{(A 7)}$$

If the matrix $\boldsymbol{B}$ is orthogonal, (A 7) expresses a similarity transformation, and the primary and modified diffusivity tensors $\boldsymbol{D}$ and $\boldsymbol{S}$ share their eigenvalues. It is worth noting that (A 4) is consistent with the general relationship $\partial F_{ij}/\partial\hat{t} = F_{kj}\,\partial v_i/\partial x_k$, where $\boldsymbol{F}$ is the deformation gradient, which is applicable to more general nonlinear flows (Goddard 1993).

Using the continuity equation, we place (A 6) into a more compact form involving the density ρ, which varies in time in response to the uniform time-dependent rate of expansion of the fluid, as

$$\frac{\partial}{\partial \hat{t}}\left(\frac{G^{CD}}{\rho}\right) = \boldsymbol{S}(t):\hat{\nabla}\hat{\nabla}\left(\frac{G^{CD}}{\rho}\right) + \frac{1}{\rho}\,\delta(x-x_0)\,\delta(y-y_0)\,\delta(z-z_0)\,\delta(\hat{t}). \qquad \text{(A 8)}$$

The solution of (A 8) is given by

$$G^{CD} = \frac{\rho}{\rho(t=t_0)}\frac{1}{(4\pi)^{3/2}}\frac{1}{\beta^{1/2}}\exp\left(-\tfrac{1}{4}\boldsymbol{J}^{-1}:\hat{\boldsymbol{x}}\hat{\boldsymbol{x}}\right), \qquad \text{(A 9)}$$

where

$$\boldsymbol{J}(t) = \int_{t_0}^{t} \boldsymbol{S}(\tau)\,\mathrm{d}\tau$$

and

$$\beta = \mathrm{Det}\,(\boldsymbol{J}) \qquad \text{(A 11)}$$

(Novikov 1958). The derivation of (A 9) uses the identity

$$\frac{\mathrm{d}}{\mathrm{d}t}\{\ln[\mathrm{Det}\,(\boldsymbol{H})]\} = \frac{\mathrm{d}\boldsymbol{H}}{\mathrm{d}t}:\boldsymbol{H}^{-1^T} = \mathrm{Tr}\left[\boldsymbol{H}^{-1}\cdot\frac{\mathrm{d}\boldsymbol{H}}{\mathrm{d}t}\right], \qquad \text{(A 12)}$$

which is valid for any non-singular differentiable square matrix function $\boldsymbol{H}(t)$ (Gradshteyn & Ryshik 1980, p. 1108). Equation (A 4), in conjection with (A 12) applied for $\boldsymbol{H} = \boldsymbol{B}$, confirms that, when velocity field is solenoidal, Det$\,(\boldsymbol{B})$ remains equal to unity at all times.

The solution of (A 4) and (A 5) with initial conditions $\boldsymbol{B}(t_0) = \boldsymbol{I}$ and $\boldsymbol{c}(t_0) = \boldsymbol{x}_0$ is given by

$$\boldsymbol{B}(t) = \exp\left[-\int_{t_0}^{t}\boldsymbol{A}(\tau)\,\mathrm{d}\tau\right] \qquad \text{(A 13)}$$

and

$$\boldsymbol{c}(t) = \boldsymbol{x}_0 + \int_{t_0}^{t}\boldsymbol{B}(\tau)\cdot\boldsymbol{U}(\tau)\,\mathrm{d}\tau. \qquad \text{(A 14)}$$

Steady flow

When $\boldsymbol{A}$ and $\boldsymbol{U}$ are constant, independent of time, (A 13) and (A 14) give

$$\boldsymbol{B}(\hat{t}) = \exp(-\boldsymbol{A}\hat{t}), \quad \boldsymbol{c}(\hat{t}) = \boldsymbol{x}_0 + [\boldsymbol{I} - \exp(-\boldsymbol{A}\hat{t})]\cdot\boldsymbol{A}^{-1}\cdot\boldsymbol{U}. \qquad \text{(A 15)}$$

When, in addition, $\boldsymbol{D}(t)$ is isotropic with time-dependent diagonal elements equal to $D(t)$, the modified diffusivity computed from (A 7) is given by the simpler form

$$\boldsymbol{S}(t) = D(t)\exp(-\boldsymbol{A}\hat{t})\cdot\exp(-\boldsymbol{A}^T\hat{t}), \qquad \text{(A 16)}$$

and the Green's function follows readily from (A 9) and (A 10).

Batchelor (1979) and Foister & van de Ven (1980) presented alternative derivations of the Green's function for steady flow for the particular case where D is constant and independent of time. Their analyses assume a Gaussian dependence for the Eulerian spatial coordinates at the outset. Bowen & Stolzenbach (1992) used their results to study the properties of the corresponding stationary Green's function describing the scalar field due to a permanent point source.

To study the behaviour of the Green's function at short times, we expand the exponential terms in (A 16) in Taylor series with respect to their arguments and obtain

$$\boldsymbol{S}(t) = D(t)\,(\boldsymbol{I} - 2\boldsymbol{E}\hat{t} + (2\boldsymbol{E}^2 + \Xi\cdot\boldsymbol{E} - \boldsymbol{E}\cdot\Xi)\,\hat{t}^2 + \ldots), \qquad \text{(A 17)}$$

where $\boldsymbol{E} = \frac{1}{2}(\boldsymbol{A} + \boldsymbol{A}^T)$ is the rate of deformation tensor, and $\Xi = \frac{1}{2}(\boldsymbol{A} - \boldsymbol{A}^T)$ is the vorticity tensor. When D is independent of time, we integrate (A 17) according to (A 10) and obtain

$$\boldsymbol{J}(\hat{t}) = D\hat{t}(\boldsymbol{I} - \boldsymbol{E}\hat{t} + \tfrac{1}{3}(2\boldsymbol{E}^2 + \Xi\cdot\boldsymbol{E} - \boldsymbol{E}\cdot\Xi)\,\hat{t}^2 + \ldots) \tag{A 18}$$

from which it follows that, for an incompressible fluid,

$$\beta = D^3\hat{t}^3(1 + \tfrac{1}{6}\boldsymbol{E}:\boldsymbol{E}\hat{t}^2 + \ldots), \tag{A 19}$$

where $\boldsymbol{E}:\boldsymbol{E} = \mathrm{Tr}\,(\boldsymbol{E}^2)$, as deduced by Batchelor (1979) using a different method. Equations (A 18) and (A 19) show that the rotation of the fluid around the point source plays a secondary role during the initial stages of transport. The rotary motion simply moves the fluid parcels over the nearly spherical iso-scalar surfaces centred at the point source.

REFERENCES

Abramzon, B. & Elata, C. 1984 Unsteady heat transfer from a single sphere in Stokes flow. *Intl J. Heat Mass Transfer* **27**, 687–695.

Acrivos, A. 1980 A note on the rate of heat or mass transfer from a small particle freely suspended in a linear shear flow. *J. Fluid Mech.* **98**, 299–304.

Acrivos, A. & Taylor, T. D. 1962 Heat and mass transfer from single spheres in Stokes flow. *Phys. Fluids* **5**, 387–394.

Batchelor, G. K. 1979 Mass transfer from a particle suspended in fluid with a steady linear ambient velocity distribution. *J. Fluid Mech.* **95**, 369–400.

Batchelor, G. K. 1980 Mass transfer from a small particle suspended in turbulent fluid. *J. Fluid Mech.* **98**, 609–623.

Bowen, J. D. & Stolzenbach, K. D. 1992 The concentration distribution near a continuous point source in steady homogeneous shear. *J. Fluid Mech.* **236**, 95–110.

Brenner, H. 1961 The Oseen resistance of a particle of arbitrary shape. *J. Fluid Mech.* **11**, 604–610.

Brenner, H. 1963 Forced convection heat and mass transfer at small Péclet numbers from a particle of arbitrary shape. *Chem. Engng Sci.* **18**, 109–122.

Brunn, P. O. 1984 Inertia effects for slow forced scalar transfer from a sphere. *Acta Mechanica* **50**, 271–284.

Chandrasekhar, S. 1943 Stochastic problems in physics and astronomy. *Rev. Mod. Phys.* **15**, 1–89.

Choudhury, P. N. & Drake, D. G. 1971 Unsteady heat transfer from a sphere in a low Reynolds number flow. *Q. J. Mech. Appl. Maths* **24**, 23–36.

Clift, R., Grace, J. R. & Weber, M. E. 1978 *Bubbles, Drops, and Particles*. Academic.

Feng, Z.-G. & Michaelides, E. E. 1996 Unsteady heat transfer from a sphere at small Péclet numbers. *Trans. ASME J. Fluids Engng* **118**, 96–102.

Foister, R. T. & Ven, T. G. M. van de 1980 Diffusion of Brownian particles in shear flows. *J. Fluid Mech.* **96**, 105–133.

Frankel, N. A. & Acrivos, A. 1968 Heat and mass transfer from small spheres and cylinders freely suspended in shear flow. *Phys. Fluids* **11**, 1913–1918.

Gibert, H. & Angelino, H. 1973 Influence de la pulsation sur les transferts de matière entre une sphère et un liquide. *Can. J. Chem. Engng* **51**, 319–325.

Goddard, J. D. 1993 The Green's function for passive scalar diffusion in a homogeneously sheared continuum. *Phys. Fluids* A **5**, 2295–2297.

Gradshteyn, I. S. & Ryzhik, I. M. 1980 *Tables of Integrals, Series, and Products*. Academic.

Kaiping, P. 1983 Unsteady forced convective heat transfer from a hot film in non-reversing and reversing shear flow. *Intl J. Heat Mass Transfer* **26**, 545–557.

Konopliv, N. & Sparrow, E. M. 1971 Temperature and heat transfer history of a solid body in a forced convection flow. *Q. Appl. Maths* **29**, 225–235.

Konopliv, N. & Sparrow, E. M. 1972 Unsteady heat transfer and temperature for Stokesian flow about a sphere. *J. Heat Transfer* **94**, 266–272.

Krishnan, G. P. & Leighton, D. T. jr 1992 Diffusion from a point source in a time-dependent unbounded extensional flow. *Phys. Fluids* A **11**, 2327–2331.

LEIGHTON, D. T. JR 1989 Diffusion from an initial point distribution in an unbounded oscillating simple shear flow. *Physicochem. Hydrodyn.* **11**, 377–386.

NOVIKOV, E. A. 1958 On turbulent diffusion in a flow with transverse velocity gradient. *Prikl. Mat. Mek.* **22**, 412–414 (In Russian).

PEDLEY, T. J. 1972 On the forced heat transfer from a hot film embedded in the wall in two-dimensional unsteady flow. *J. Fluid Mech.* **55**, 329–357.

PEDLEY, T. J. 1976 Heat transfer from a hot film in reversing shear flow. *J. Fluid Mech.* **78**, 513–534.

POLYANIN, A. D. 1984 An asymptotic analysis of some nonlinear boundary-value problems of convective mass and heat transfer of reacting particles with the flow. *Intl J. Heat Mass Transfer* **27**, 163–189.

POZRIKIDIS, C. 1989 A singularity method for unsteady linearized flow. *Phys. Fluids* A **1**, 1508–1520.

TOWNSEND, A. A. 1960 The diffusion of heat spots in isotropic turbulence. *Proc. R. Soc. Lond.* A **209**, 418–430.