A statistical model of fluid-element motions and vertical diffusion in a homogeneous stratified turbulent flow

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The vertical displacements Z(t) of fluid elements passing through a source z = 0 at t = 0 in a horizontal mean flow with stably stratified statistically stationary turbulence (with buoyancy frequency N and velocity time-scale T), under the action of random pressure gradients and damping by internal wave motions, are investigated by a model Langevin-like equation, and by a general Lagrangian analysis of the displacements, of the density flux and of the energy of fluid elements. Solutions for the mean-square displacement $\overline{Z}^2(t)$, the mean-square velocity \overline{w}^2 , and the autocorrelation of the velocity are calculated in terms of the spectrum $\Phi(s)$ of the pressure gradient. We use model equations for the momentum of fluid elements and for the exchange of density fluctuations between fluid elements, taking the elements' diffusion timescale to be γ^{-1} times the buoyancy timescale N^{-1} , where γ is a measurable parameter.

In the case of moderate-to-strong stable stratification (i.e. $NT \gtrsim 1$), we find the following.

(i) When there is no change of the fluid elements' density ($\gamma = 0$), the mean-square displacement $\overline{Z^2}$ of marked particles ceases to grow when $t \gtrsim N^{-1}$, and its asymptotic value is proportional to $\overline{w^2}/N^2$, where the constant of proportionality, $\overline{\zeta^2}(\infty)$, is O(1) and a decreasing function of $(NT)^{-1}$. This result is also shown to be a general consequence of the finite potential and kinetic energy in the stationary turbulence.

(ii) If there is a small diffusive interchange of density between fluid elements (i.e. $\gamma \ll 1$), the marked particles' mean-square displacement has a slow linear growth (i.e. $\overline{Z^2} \sim \overline{w^2}/N^2(1+O(\gamma^2)tN)$.

(iii) Such molecular processes must also dilute the initial concentration of contaminants (e.g. dye or smoke) in those fluid elements that diffuse above the limit in (i).

(iv) The mean-square fluctuation of density is proportional to the product of the asymptotic mean-square displacement of marked particles and the square of the mean density gradient $(-\mathscr{G})$ (i.e. $\overline{\rho'^2} = \mathscr{G}^2 \overline{Z^2} = \frac{1}{2} \overline{\zeta^2}(\infty, \gamma = 0) \, \mathscr{G}^2 \overline{w^2}/N^2$).

(v) The flux F_{ρ} of density in a turbulent flow can be expressed exactly as the sum of two terms, the first $\frac{1}{2}(d\overline{Z}^2/dt) \mathscr{G}$, being caused by the growth of the displacements of fluid elements, and the second $\overline{Z}(\rho + \rho')$ being caused by the mixing between fluid elements. In stable flows, it is shown that the second element is dominant, and $F_{\rho} \sim \gamma \overline{w^2} N \rho_0 g$ while the first is smaller by $O(\gamma)$.

Previous laboratory and field measurements of $\overline{w^2}$, $\overline{Z^2}$, $\overline{\rho'^2}$ and $R_W(t)$ are discussed in some detail and shown to be consistent with this model.

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1. Introduction

The vertical diffusion of particles released from a steady source in a homogeneous stably stratified turbulent flow is poorly understood, even at a qualitative level. From a practical point of view, this lack of physical understanding means that even the appropriate dimensionless form of the relationship between the vertical displacement Z and the statistical features of the vertical velocity field w are uncertain. We list below some of the salient features of vertical diffusion with some of the questions that they raise about the diffusion and the dynamics.

(i) Consider a turbulent flow with neutral stability. On average, fluid elements from a source move vertically an ever increasing distance as time proceeds, while the mean-square fluctuating velocity of a fluid element remains constant (Lin & Reid 1963). However, in a stratified flow, a large source of energy is required to give fluid elements large vertical displacements (assuming their densities remain unchanged). For a given kinetic energy of fluid elements (equal to $\frac{1}{2}\rho \overline{w^2}$, where $\overline{w^2}$ is the mean-square vertical velocity as measured at a point), and a roughly equal partition of energy between kinetic and potential forms, it follows that fluid elements can only diffuse through a vertical distance of the order $\overline{(w^2)^2}/N$, where $N = [g\rho_0^{-1} \mathscr{G}]^{\frac{1}{2}}, \rho_0$ is the mean density, g is the gravitational acceleration and $-\mathcal{G}$ is the mean density gradient, which is assumed to be uniform and is not affected by turbulence. So, to understand vertical diffusion in stable conditions, one must consider the dynamical effects of buoyancy forces (Priestley 1959). One must also consider mixing because, over sufficiently long times, there is always some exchange of density between elements of the fluid by molecular processes, and then the vertical diffusion can increase without limit. Under what circumstances and over what timescales do these density changes become more significant? If the density must change by molecular diffusion for fluid particles to travel large distances, then surely the concentration of contaminant marking the fluids must be reduced by molecular diffusion?

(ii) An important practical question is whether such plume behaviour is consistent with representing the diffusion by a gradient transport model, involving an eddy diffusivity. Consider, at a time t after their 'release' or their 'marking', the mean-square vertical displacement \overline{Z}^2 of an ensemble of fluid elements. If the Lagrangian autocorrelation function of the vertical velocity W(t) of the elements is $R_W(\tau) = \overline{W(t) W(t+\tau)}$, then, following Taylor (1921), when $t \gg T_L$

$$\overline{Z^2} \sim 2 \overline{W^2} \bigg[T_{\rm L} t - \int_0^\infty \tau R_W(\tau) \, d\tau \bigg], \tag{1.1}$$

where

$$T_{\rm L} = \int_0^\infty R_W(\tau) \, d\tau, \quad \overline{W^2} = \overline{w^2}. \tag{1.2}$$

The second term of (1.1) is negligible, if $T_{\rm L} \neq 0$ as $t/T_{\rm L} \rightarrow \infty$, so that $\overline{Z^2}$ grows linearly with time as if it were governed by a diffusion equation with diffusivity $\overline{w^2}T_{\rm L}$. But if $d\overline{Z^2}/dt \approx 0$ at large times, as in the laboratory experiments of Britter *et al.* (1983 hereinafter referred to as BHMS), and some of the field observations of Hilst & Simpson (1958) and Koføed-Hansen (1962), then it appears as if $T_{\rm L} \approx 0$ and the second term dominates, clearly a most undiffusionlike process!

(iii) This raises the question of how $R_W(t)$ changes its form as the timescale of vertical oscillation in a stratified fluid becomes smaller than the dynamical 'turnover' or Lagrangian timescale $T \,(\approx L_x/(\overline{w^2})^{\frac{1}{2}})$, i.e. as NT increases $(L_x$ is the spatial integral

of the vertical velocity fluctuations). Some laboratory measurements of $R_W(t)$ by Frenzen (1963) will be compared with our theory.

(iv) In a stably stratified turbulent flow, not only does the vertical velocity at a point vary randomly, but so does the density. Therefore there may be a difference between the behaviour of an ensemble of fluid elements of one density and that of all fluid elements passing a point. Which of these two ensembles represents dye or smoke of constant density released into a turbulent flow? Another implication of the density fluctuations is that it may be possible to infer, from measurements of fluctuating density at a point, the vertical diffusion of particles released into a flow (Lange 1974).

These features of turbulent diffusion in a stably stratified flow can only be understood by a dynamical and kinematical analysis of the vertical motion of fluid elements whose density can change by mixing. Since we are particularly interested in diffusion a Lagrangian analysis is essential, but, because it is a Lagrangian rather than an Eulerian analysis, it is inevitably more speculative. Most approximate models of stratified turbulence have been developed in an Eulerian framework for the purpose of estimating mean flows, scalar fluxes and variance of turbulent velocities (e.g. Launder 1976).

In the Lagrangian mathematical model developed in this paper, the turbulence is assumed to be homogeneous and stationary in time. In reality turbulence must either be slowly decaying (as in homogeneous turbulence in the nocturnal layer), or sustained by shear or interactions of internal waves. In the former case we may assume that the timescale of decay is somewhat greater than the buoyancy timescale N^{-1} (see §5). In the latter case we assume that the vertical fluctuations are stationary random functions of time (see §4.3).

The model is an extension of that of Csanady (1964), and is of the Langevin or 'random-force' type used previously to describe turbulent diffusion in neutral flows (Lin & Reid 1963; Krasnoff & Peskin 1971), and similar in form to the Langevin equations in the theory of Brownian motion. The aim of such models is to derive the vertical displacement $\overline{Z^2(t)}$ and the Lagrangian velocity autocovariance $\overline{W^2}R_W(t)$ of small particles in terms of some random forces acting on them. Here we consider fluid elements under the action of pressure gradient fluctuations. The theory is only useful if $\overline{Z^2}$ and $R_W(t)$ can be deduced from $\overline{W^2}$ and from weak assumptions about the pressure-gradient spectrum $\Phi(s)$. A useful deductive theory can be constructed firstly because $\Phi(s)$ is essentially a 'white-noise' spectrum over most of the inertial subrange of spectrum (Monin & Yaglom 1975, §18.3) and secondly because $\Phi(s)$ is largely determined by the rate of energy dissipation per unit mass ϵ , which is insensitive (for given $\overline{w^2}$ and T) to stratification (see BHMS and §2.3). If the change of a fluid element's density is estimated by Burgers' model (Hinze 1975), then the model also shows how the density flux varies with N, $\overline{w^2}$ and T.

The model equations may be viewed in two equivalent ways, either as derived from the exact equations of motion of the fluid in a Lagrangian form by the approximation of certain terms, or in a more physically appealing way as idealized equations for the motion of finite but small parcels of fluid, with dimensions small compared with any integral scale of the turbulence, but large compared with whichever is the larger of the viscous Kolmogorov microscale, and the diffusive microscale, defined in Monin & Yaglom (1975, p. 200).

In Csanady's (1964) theory, the pressure gradient and viscous forces on a fluid element are entirely replaced by a stochastic force; in contrast, Lin & Reid (1963) and Krasnoff & Peskin (1971, hereinafter referred to as KP) add a damping term, linear in the velocity, but which is primarily intended to represent the effects of viscosity. In a stable density gradient there can be wave-like motions at all scales, and we suggest that the generation and transmissions of such motions act as a local damping mechanism for vertical motions of the fluid. Therefore this damping is approximately proportional to the (local) buoyancy frequency. The model has both turbulent and wave-like features without drawing a sharp distinction between them. Further discussion of this point is given by BHMS. The observations of Gartrell (1979) and Hunt *et al.* (1982), for instance, show clearly how motions in a stably stratified *shear* flow can be a mixture of waves and turbulence.

In §2 the basic model is set up, the equations solved, and the form of the random forcing discussed. In §3 numerical and analytic results for plume shapes, vertical velocity correlations and density fluxes are presented. In §4 the relation between density exchange among fluid elements and the vertical density flux through the system is investigated, both in terms of the model and by more general arguments. In §5, the theory is compared with experimental observations, in particular the recent results of BHMS on plume shapes and velocity fluctuations in stratified grid turbulence. An attempt is made to correct the theory for decaying turbulence intensities.

2. A random-force model of fluid-element motions

2.1. Equations of motion

If W is the vertical velocity, ν the kinematic viscosity, κ the molecular diffusivity of the scalar responsible for the density gradient, ρ' the density perturbation from the local mean value ρ , p' the pressure perturbation from its hydrostatic value, then, on making the Boussinesq approximation, the equations for perturbation quantities are, following a fluid element (Csanady 1964, equations (6), (11)),

$$\frac{dW}{dt} + \frac{g\rho'}{\rho_0} = -\frac{1}{\rho_0} \frac{\partial p'}{\partial z}(t) + \nu \nabla^2 W(t), \qquad (2.1)$$

$$\frac{d(\rho+\rho')}{dt} = \frac{d\rho'}{dt}(t) - W(t) \mathcal{G} = \kappa \nabla^2 \rho'(t).$$
(2.2)

In the absence of molecular diffusion W(t) defines the velocity of a vanishing small volume of *matter*. But since molecular diffusion enables molecules to pass in and out of this volume, eventually it may contain none of the original molecules, and therefore W is defined over a vanishing small 'control volume' moving at the local fluid velocity (Saffman 1960) \dagger – figure 1 (a). A small but finite control volume loses even the original fluid elements (this takes a timescale $t \sim e^{-\frac{1}{2}l_0^2}$ if l_0 is small, but larger than the microscale $(v^3/e)^{\frac{1}{2}}$). In this case the limit W describes the motion of a small material volume only over a limited time. The eventual difference between molecular trajectories and fluid element trajectories makes the deduction of the latter from *concentration* measurements somewhat conjectural (figure 1b)! Fluid elements which pass through the source are referred to as 'marked fluid elements', even though it is likely that any *physical sign* of their marking may transfer itself to other fluid elements.

In considering how to model the damping of vertical motions by viscous forces and the generation and propagation of waves, we recall two experiments. In the first a

[†] It was pointed out by a referee that this implies that $d\rho/dt \neq 0$ and therefore that $\nabla \cdot \mathbf{u} \neq 0$. However, the error on scales $O(\rho_0/\mathscr{G})$ is negligible.



FIGURE 1. (a) Movements of a small patch of pollutant released from source: ---, paths of marked molecules or particles of source (e.g. dye or smoke); ---, fluid-element paths (parallel to local velocity vector). Note how they eventually diverge. (b) Regions of plume behaviour in typical plume in stable condition: (A) source region where contaminant diffuses into the fluid and is accelerated into motion; (B) fluid element and marked molecules move together; (C) divergence of paths between fluid elements and marked molecules.



FIGURE 2. Forces on a fluid element at Z(t) moving upwards with a velocity W(t). $Z_0(t)$ is a typical trajectory of an element released above its equilibrium point Z_e with no ambient turbulence.

ball is released in a stratified tank of water above or below its equilibrium level and returns to its equilibrium position in one or two oscillations, as if the fluid is as viscous as treacle (see, e.g. the film by Hunt (1976) – figure 2). Larsen's (1969) experiments and calculations of a sphere oscillating about its position of equilibrium showed that this 'stickiness' is really due to the sphere radiating waves with most of the viscous dissipation taking place far away. He calculates an exact representation for smallamplitude motions of the sphere

$$\frac{d^2Z}{dt^2} + N^2Z = \frac{Z(t) - Z(0)}{t^2} - \frac{1}{t}\frac{dZ}{dt}$$

A simpler but less accurate representation is

$$\frac{d^2Z}{dt^2} + \beta \frac{NdZ}{dt} + N^2 Z = 0, \qquad (2.3)$$

if the dimensionless parameter β (\leq 1) is taken to be a slowly varying function of amplitude. In a second experiment, a parcel of *fluid* is released away from its equilibrium level in a linear density gradient (Cerasoli 1978), and a similar damped motion is observed. Cerasoli's experiments suggest that most energy is propagated away at an angle $\frac{1}{4}\pi$ to the horizontal, corresponding to a frequency of oscillation of the entraining fluid parcel of $\sqrt{\frac{1}{2}} N$. This corresponds to a value of $\beta = \sqrt{\frac{1}{2}} = 0.7$ in (2.3). (Note that the speed of propagation of such internal waves in a turbulent flow is of order NL_x . This is much less than the mean-flow velocity, since $NL_x \sim \sigma_W \ll U$.)

On the basis of these observations we assume that the pressure and viscous forces acting on a fluid element in a turbulent flow can be modelled by

$$-\frac{1}{\rho_0}\frac{dp'}{dz} + \nu \nabla^2 W(t) = -2\beta N W(t) + H(t), \qquad (2.4)$$

where the damping coefficient βN is approximately linear in N. We do not see much merit in refining (2.4), at this stage of our understanding, to allow the damping to be frequency-dependent, e.g. by replacing $-2\beta W(t)$ in (2.4) by a convolution integral

$$-2\int_{\infty}^{t}\mu(t-\eta) W(\eta) d\eta,$$

for some damping function μ .

As in previous models the fluctuating pressure gradient H(t) is assumed to be a stationary random function. In fact this model for the pressure and viscous forces on a fluid element can be thought of as representing the forces due to the wave motions set up by *that fluid element* in the first term and the forces due to the accelerations and waves caused by *all other fluid elements* in the second term H(t).

The rate of change of the density difference between a fluid element and the mean density of its environment is determined by its vertical advection and by molecular diffusion acting on local density gradients. It is natural to express the timescale for this process in terms of N. Following Burgers (see Hinze 1975, chap. 5) and Csanady (1964), we model the density diffusion by

$$k\nabla^2 \rho' = -\gamma N \rho', \qquad (2.5)$$

where $(\gamma N)^{-1}$ is the timescale for the fluid elements to change their density. There is now evidence to suggest that γ is only a slowly varying function of NT, where T is a typical timescale of the turbulence. See §4 and Hunt *et al.* (1982) for discussion of the value of γ .

Combining (2.1), (2.2), (2.4) and (2.5) gives

$$\frac{dW}{dt} + 2\beta NW + \frac{g\rho'}{\rho_0} = H(t), \qquad (2.6)$$

$$\frac{d\rho'}{dt} - \mathscr{G}W = -\gamma N\rho'. \tag{2.7}$$

These are the model equations for the position and density of a fluid element, which are solved in §2.2.

2.2. Method of solution

The coupled linear stochastic equations (2.6), (2.7) are solved by Fourier-transform methods to find $\overline{W^2}(t)$, $\overline{Z^2}(t)$ and $\overline{\rho'^2}(t)$ in terms of the spectrum $\Phi(s)$ of H(t), which is assumed to be a stationary random function. Defining

$$H(t) = \int_{-\infty}^{\infty} e^{ist} \hat{H}(s) \, ds; \qquad (2.8a)$$

Vertical diffusion in stratified turbulent flow 225

where

$$\hat{H}(s')\,\hat{H}^*(s) = \delta(s-s')\,\Phi(s), \qquad (2.8b)$$

then W(t) and $\rho'(t)$ can also be expressed in terms of $\hat{H}(s)$ by means of the deterministic transfer functions M(s, t) and Q(s, t) as

$$W(t) = \int_{-\infty}^{\infty} M(s,t) \,\hat{H}(s) \, ds, \quad \rho'(t) = \int_{-\infty}^{\infty} Q(s,t) \,\hat{H}(s) \, ds. \tag{2.9}$$

Substitution of (2.8) and (2.9) into (2.6) and (2.7) leads to the equations for M, Q:

$$\frac{dM}{dt} + 2\beta NM + \frac{gQ}{\rho_0} = e^{ist}, \qquad (2.10)$$

$$\frac{dQ}{dt} - \mathscr{G}M = -\gamma NQ. \tag{2.11}$$

Since W(t) and $\rho'(t)$ must also be stationary random functions, the appropriate solutions to (2.10), (2.11) are

$$W(t) = \int_{-\infty}^{\infty} \frac{(\gamma N + is) \hat{H}(s) e^{ist}}{E(s)} ds, \qquad (2.12)$$

$$\rho'(t) = \mathscr{G} \int_{-\infty}^{\infty} \frac{\hat{H}(s) e^{ist}}{E(s)} ds, \qquad (2.13)$$

where

$$E(s) = N^2(1+2\beta\gamma) - s^2 + iN(2\beta+\gamma) s.$$

To evaluate Z(t), we need only integrate (2.12) with respect to time. The constant of integration is supplied by the constraint that the fluid element passes the origin at zero time, i.e. Z(0) = 0 for each realization. Then

$$Z(t) = \int_{-\infty}^{\infty} \frac{((\gamma N/is) + 1) \hat{H}(s)}{E(s)} (e^{ist} - 1) \, ds, \qquad (2.14)$$

which is the vertical displacement of a fluid element whose density and velocity at t = 0 are random. It may be of interest to calculate $\overline{Z^2}(t)$ for the ensemble of fluid elements whose equilibrium level is equal to the source height, i.e. $\rho'(t=0) = 0$. Solving (2.6), (2.7) subject to this initial condition (in the limit $\gamma = 0$) yields

$$Z(t) = \int_{-\infty}^{\infty} \frac{\hat{H}(s)}{E(s)} \left\{ e^{ist} - e^{-\beta Nt} \left[\cos N\delta t + \frac{\beta}{\delta} \sin N\delta t \right] \right\} ds,$$
(2.15)

where $\delta = (1 - \beta^2)^{\frac{1}{2}}$ ($\beta < 1$ by assumption).

Whether a real plume is more accurately modelled as the passive marking of fluid elements of all densities, as they pass the source, or alternatively by only averaging over an ensemble of elements of fixed density depends on the details of any initial mixing very close to the source (Puttoek 1976).

2.3. Computation of variances

The variances $\overline{W^2}(t)$, $\overline{Z^2}(t)$, $\overline{\rho'^2}(t)$ and the velocity autocorrelation function $R_{W}(t) = \overline{W(t) W(t+\tau)}/\overline{W^2}$ are now calculated in terms of the normalized pressuregradient spectrum

$$\tilde{\Phi}(\omega) = \Phi(s) / \overline{W^2} N, \qquad (2.16)$$

where $\omega = s/N$.

The weak effect of stratification on $\Phi(s)$ depends on the parameter $\alpha = 1/(NT)$.



FIGURE 3. Sketches of the Lagrangian pressure-gradient spectra $\tilde{\Phi}^{(a)}(\omega)$ and $\tilde{\Phi}^{(b)}(\omega)$ corresponding to forms of (2.25a, b), (when $\alpha Re^{\frac{1}{2}} \gg 1$, $\gamma \ll 1$), and of the transfer functions $\tilde{P}^{-1}(\omega)$ and $\omega^2 \tilde{P}^{-1}$ relating mean-square displacement $\overline{Z^2}$ and velocity $\overline{W^2}$ to the pressure-gradient spectrum $\tilde{\Phi}(\omega)$, when $Nt \gg 1$ and $\gamma^2 Nt \ll 1$.

By standard methods (e.g. Yaglom 1962) for stationary random processes, it follows from (2.12)-(2.14) that

$$R_{W}(\tau) = \int_{-\infty}^{\infty} \frac{(\gamma^{2} + \omega^{2}) \,\tilde{\Phi}(\omega, \alpha) \, e^{i\omega N\tau}}{\tilde{P}(\omega)} d\omega, \qquad (2.17)$$

$$\overline{\rho'^2} = \mathscr{G}^2 \overline{W^2} N^{-2} \int_{-\infty}^{\infty} \tilde{\Phi}(\omega, \alpha) \, \tilde{P}^{-1}(\omega) \, d\omega, \qquad (2.18)$$

where

$$\widetilde{P}(\omega) = (1 + 2\beta\gamma - \omega^2)^2 + (\gamma + 2\beta)\,\omega^2, \qquad (2.19a)$$

and $\Phi(\omega, \alpha)$ satisfies the normalization condition

$$1 = \int_{-\infty}^{\infty} (\gamma^2 + \omega^2) \, \tilde{\Phi}(\omega, \alpha) \, \tilde{P}^{-1}(\omega) \, d\omega.$$
 (2.19b)

Also $\overline{Z^2}(t) = \overline{\zeta^2}(Nt) \overline{W^2}/N^2$, where, if the ensemble of *all* particles passing through the source is considered,

$$\overline{\zeta^2}(Nt) = 2 \int_{-\infty}^{\infty} \left(1 + \frac{\gamma^2}{\omega^2}\right) \widetilde{P}^{-1}(\omega) \left(1 - \cos \omega t N\right) \widetilde{\Phi}(\omega, \alpha) \, d\omega.$$
(2.20)

See figures for sketches of typical forms of \tilde{P}^{-1} and $\tilde{\Phi}$.

Since the turbulence is homogeneous and stationary, Lumley's (1962) argument shows that the ensemble or time mean-square velocity of a fluid element should be equal to that of all fluid elements and should also be equal to the mean-square velocity measured at any point in the flow, i.e.

$$\overline{W^2} = \overline{w^2}.\tag{2.21}$$

In strongly stratified flows, the velocity integral timescale can be rather anomalous as γ or N vary. If it is defined as usual, then from (2.17)

$$T_{\rm L} = \int_0^\infty R_W(\tau) \, d\tau = \frac{\pi N^{-1} \gamma^2 \Phi(\omega = 0)}{(1 + 2\beta\gamma)^2}.$$
 (2.22*a*)

Thus $T_{\rm L} = 0$ if $\tilde{\Phi}(\omega = 0) = 0$ or if $\gamma \to 0$ (i.e. no mixing occurs). Therefore it may be convenient to define another integral scale $T_{\rm L}^{(1)}$ in terms of the first moment of $R_W(\tau)$

$$T_{\rm L}^{(1)} = \left| \int_{0}^{\infty} \tau R_{W}(\tau) \, d\tau \right|^{\frac{1}{2}} = N^{-1} \left| \int_{-\infty}^{\infty} \frac{(\omega^{2} + \gamma^{2}) \, \tilde{\Phi}(\omega) \, d\omega}{\omega^{2} \tilde{P}(\omega)} \right|^{\frac{1}{2}}$$
(2.22b)
(When $\gamma = 0, \ T_{\rm L}^{(1)} = N^{-1} \overline{\zeta^{2}(\infty, \gamma = 0)} = \overline{(Z^{2})^{\frac{1}{2}}} / \overline{(W^{2})^{\frac{1}{2}}} \text{ from } (3.3b)$).

2.4. Forms of the pressure-gradient spectrum $\Phi(s)$

Since the stratification affects Z and W it must also affect $\Phi(s)$. It is certainly observed in the atmospheric boundary layer that $\Phi(s)$ is changed by stratification in the energy-containing range (i.e. $s \leq \sigma_W/L_x$). However, it is also observed that the effects of even moderately stable (or unstable) stratification do not measurably change the structure of the intertial subrange of the Eulerian turbulent velocity field, nor the value of the Kolmogorov constant in the inertial subrange, nor the value of $\epsilon/((\overline{w^2})^{\frac{2}{3}}/L_x^{(w)})$ where $L_x^{(w)}$ is the integral scale of the vertical velocity fluctuations. (By moderately we mean $(\overline{w^2})^{\frac{1}{2}}/(L_x^{(w)} N) \gtrsim 1$; see e.g. Kaimal 1973; BHMS; Hunt *et al.* 1982.)

In neutral conditions in the inertial subrange, the Lagrangian velocity spectrum is (Monin & Yaglom 1975, p. 361)

$$\Phi_{W}(s) = B_0 \epsilon s^{-2}, \qquad (2.23)$$

where B_0 is a universal constant ($B_0 \approx 0.6$, according to recent atmospheric observations by Hanna 1981). Consequently in neutral conditions, when $sT \ge 1$, since the pressure-gradient spectrum must be equal to the product of the acceleration spectrum (Monin & Yaglom 1975b, p. 371) and ρ_0^2 ,

$$\Phi(s) \propto \epsilon, \quad \tilde{\Phi}(\omega, \alpha) \sim \operatorname{const} \propto \epsilon/(\overline{w^2}N),$$
(2.24)

where $T \sim \epsilon/\overline{w^2} \sim T_{\rm L}^{(1)}$ and is the natural integral timescale of the turbulence. Since (2.23) is also valid in most stable conditions, we assume (2.24) is also valid.

Since a wide variety of forms of the Eulerian vertical velocity and temperature spectra are observed, we explore the implications of our model by considering two forms for $\Phi(s)$. Our results when normalized in terms of $\overline{w^2}$ are found to be rather insensitive to the forms of $\Phi(s)$, which are assumed to be as follows. (i) *Csanady's form*

$$\tilde{\Phi}^{[a]} \propto \omega^2 / (\omega^2 + \alpha^2), \qquad (2.25a)$$

which satisfies (2.24) when $\omega/\alpha \ge 1$ and tends to zero when $\omega \to 0$. It does not describe the decrease of Φ in the microscale part of the spectrum. (ii) The Krasnoff & Peskin spectrum

$$\tilde{\Phi}^{[b]} \propto \frac{1}{\omega^2 + \hat{\alpha}^2} \quad \text{where} \quad \hat{\alpha} = \alpha R e^{\frac{1}{2}},$$
(2.25b)

which satisfies (2.24) when $\omega/(\alpha Re^{\frac{1}{2}}) \ll 1$ (where $Re = (\overline{w^2})^2/\nu\epsilon$) and is finite when $\omega \to 0$. To satisfy (2.19b), when $\alpha Re^{\frac{1}{2}} \gg 1$ and, $\gamma^2 \ll 1$, in the limit of $\omega \ll Re^{\frac{1}{2}}$,

$$\tilde{\Phi}^{[b]} = 2\beta/\pi. \tag{2.25c}$$

Another spectrum with a similar form is

$$\Phi^{[b]} \propto \exp\left(-|\omega|/\hat{\alpha}\right). \tag{2.25d}$$

It is clear from (2.22) that the zero-frequency limit of Φ determines the long-term diffusion of fluid elements (or Lagrangian points in the fluid, following the continuum

velocity fluctuations). The two forms [a], [b] above for $\mathbf{\Phi}$ used respectively by Csanady and Krasnoff & Peskin have different low-frequency limits and quite different characteristic timescales (see the appendix).

3. Development of the particle displacements

3.1. Initial development

Consider the growth of $\overline{Z^2}(t)$ immediately after the release or marking of the particles. If we take the limits $t \ll N^{-1}$ and $t \ll T$, then (2.20) gives

$$\overline{\zeta^{2}}(Nt) = Nt^{2} \int_{-\infty}^{\infty} (\gamma^{2} + \omega^{2}) \widetilde{\Phi}(\omega) \widetilde{P}^{-1}(\omega) d\omega, \qquad (3.1)$$

whence $\overline{Z^2} = \overline{W^2}t^2 = \overline{w^2}t^2$ when $tN \to 0$, $t \ll T$. This is the same as Taylor's (1921) result for the initial period of diffusion when fluid-element velocities are well correlated with their initial values. This result enables $\overline{w^2}$ to be deduced from measurements of $\overline{Z^2}$ even in a stratified flow (BHMS).

3.2. Ultimate development

To obtain its asymptotic form as $Nt \rightarrow \infty$ using standard methods (e.g. Pasquill 1974, chap. 3), (2.20) is rewritten as

$$\overline{\zeta}^{2}(tN) = 2 \int_{-\infty}^{\infty} \frac{\tilde{\Phi}(\omega)}{\tilde{P}(\omega)} \left(1 - \cos \omega N t\right) d\omega + \gamma^{2} \int_{-\infty}^{\infty} \frac{\tilde{\Phi}(\omega)}{\tilde{P}(\omega)} \left(\frac{\sin\left(\frac{1}{2}\omega N t\right)}{\frac{1}{2}\omega}\right)^{2} d\omega.$$
(3.2)

Noting that $\sin \left(\frac{1}{2}\omega Nt\right)/(\frac{1}{2}\omega) \to 2\pi\delta(\omega)$ as $Nt \to \infty$, it follows, in the limits $Nt \to \infty$ and $\gamma \ll 1$, that $\overline{R}(tN \to \infty) = [\overline{R}(tN \to \infty) - [\overline{R}(tN \to \infty)] = [\overline{R}(tN \to \infty) - [\overline{R}(tN \to \infty)] = [\overline{R}(tN \to \infty)]$

$$\zeta^2(tN \to \infty) = [\zeta^2(\infty, \gamma = 0) + 2tN(NT_{\rm L})], \qquad (3.3a)$$

where

$$\overline{\zeta^{2}}(\infty,\gamma=0) = 2 \int_{-\infty}^{\infty} \tilde{\Phi}(\omega) \tilde{P}^{-1}(\omega) \, d\omega, \qquad (3.3b)$$

and

$$NT_{\rm L} = \gamma^2 \pi \lim_{\omega \to 0} \left(\frac{\tilde{\Phi}(\omega)}{\tilde{P}(\omega)} \right). \tag{3.3c}$$

Thus in the limit $\gamma \to 0$, the 'plume' (i.e. the r.m.s. displacement $(\overline{Z^2})^{\frac{1}{2}}$) levels out at a thickness $O((\overline{w^2})^{\frac{1}{2}}/N)$. But if γ is finite it grows parabolically, as in uniform neutrally stable turbulent flows. Which of these two developments occurs depends on the square of the ratio γ of the buoyancy timescale N^{-1} to the timescale for mixing between fluid elements. We shall show in §4 that γ can be estimated from the heat or density flux in a stably stratified flow. (Typically γ lies in the range 0.1–0.4 in the stable atmospheric boundary layer.)

The level of density fluctuations can be expressed in terms of the asymptotic r.m.s. displacement. From (2.18), when $\gamma \ll 1$

$$\overline{\rho'^2} = \frac{1}{2} \mathscr{G}^2 \overline{w^2} N^{-2} (\overline{\zeta^2}(\infty, \gamma = 0)) (1 + O(\gamma))$$
(3.4*a*)

$$= \frac{1}{2} \mathscr{G}^2(\overline{Z^2}(\infty, \gamma = 0) + O(\gamma \overline{w^2}/N^2)).$$
(3.4b)

In general the fluid elements, which are 'marked' as they pass through the source at Z = 0, have densities different from that of the mean density at the source, and therefore have 'equilibrium levels' which differ from Z = 0. What is the variance of



vertical positions

FIGURE 4. Fluid elements starting at equilibrium set into motion and then passing through the source S. $(\overline{Z^2})^{\frac{1}{2}}$ is plume depth of *all* particles through S; $(\overline{Z^2}^{(q)})^{\frac{1}{2}}$ is the plume depth of those particles with equilibrium height at z = 0 (denoted by dashed arrows).



FIGURE 5. Vertical-velocity autocorrelation $R_W(\tau)$. [a] From (3.6): $\beta = 0.8$, $\alpha (= 1/NT) = 1$; $\gamma = 0$. [b] From (3.7): $\beta = 0.8$. ---, typical form for $R_W(\tau)$ in neutral conditions. Note $T_L(=\int_0^\infty R_W(\tau) d\tau) = 0$ for [a] and $T_L > 0$ for [b].

the displacement of fluid elements about their equilibrium position, i.e. where $\rho'(0) = 0$? Denoting this variance by $\overline{Z^{2}(q)}$, it follows from (2.14) that

$$\overline{Z^{2}}^{(q)}(Nt \to \infty) = \overline{w^2} N^{-2} \int_{-\infty}^{\infty} \tilde{\Phi}(\omega) \, \tilde{P}^{-1}(\omega) \, d\omega. \tag{3.5a}$$

Comparing (3.5) with (3.3b), we note that

$$\overline{Z^{2}}^{(q)}(Nt \to \infty) = \frac{1}{2}\overline{Z^{2}}(\infty)$$
(3.5b)

in the absence of molecular diffusion. Thus the variance of equilibrium levels of particles being marked at the source is also $\frac{1}{2}\overline{Z^2}(\infty)$. See figure 4, and other graphs from computations in Puttock (1976).

3.3. Intermediate times ($\gamma = 0$)

For $NT \leq 1$, we would expect there to be three identifiable power-law regions of 'plume growth' or of $\overline{Z^2}(t)$. Firstly for small times $(t \leq T)$ we have the linear growth of $(\overline{Z^2})^{\frac{1}{2}}$ (3.1). In the intermediate range $(T \leq t \leq N^{-1})$, while buoyancy forces have not yet taken effect in limiting $\overline{Z^2}$, one expects the parabolic growth of $(\overline{Z^2})^{\frac{1}{2}}$ found in neutral flows at large times. Computations of the integral in (2.20) shown in figure 6(a), show this parabolic growth of $(\overline{Z^2})^{\frac{1}{2}}$. Finally, for $t \geq N^{-1}$, the analysis of §3.2 is applicable. When $NT \gtrsim 1$, there are only two regimes of the plume growth, the small-time linear growth of (3.1) merging into the asymptotic constant 'plume' depth given by (3.3b).

3.4. Analytic and numerical evaluations

The two forms of the pressure-gradient spectra specified in (2.25a, b) yield different expressions for the autocorrelation function $R_W(\tau)$, and for the growth of $\overline{Z^2}$, expressed in the dimensionless form $\overline{\zeta^2}$: (a)

$$R_{W}^{[\alpha]}(\tau) = \frac{2\alpha^{3}\beta e^{-\alpha\tau N} + \delta^{-1} e^{-\beta\tau N} \left[\delta(1+\alpha^{2}(1-4\beta^{2})\cos\delta\tau N) - \beta(1+\alpha^{2}(3-4\beta^{2})\sin\delta\tau N)\right]}{B(1+2\alpha\beta)}, \\ \overline{\zeta}^{2^{[\alpha]}}(\tau) = \frac{\left[2B+4\alpha\beta e^{-\alpha\tau N} - 2\delta^{-1} e^{-\beta\tau N} (\delta(1+\alpha^{2})\cos\delta\tau N + \beta(1-\alpha^{2})\sin\delta\tau N)\right]}{B(1+2\alpha\beta)},$$

$$(3.6)$$

where

$$\delta = (1 - \beta^2)^{\frac{1}{2}}, \quad B = 1 - 2\alpha\beta + \alpha^2;$$

$$-\delta^{-1}e^{-\beta tN}(\delta(1-4\beta^2+\hat{\alpha}^2)\cos\delta tN+\beta(3-4\beta^2+\hat{\alpha}^2)\sin\delta\tau N)],$$

where

The asymptotic mean-square displacements (for $\gamma = 0$) are

(a)
$$\overline{\zeta^2}(\infty, \gamma = 0)^{[\alpha]} = \frac{2}{1 + 2\alpha\beta}, \qquad (3.8)$$

 $\hat{B} = 1 - 2\hat{\alpha}\beta + \hat{\alpha}^2.$

(b)
$$\overline{\zeta^2}(\infty, \gamma = 0)^{[b]} = 2\left(1 + \frac{2\beta}{\alpha R e^{\frac{1}{2}}}\right). \tag{3.9}$$

Since $Re \ge 1$, spectrum [b] reduces to const/ $\hat{\alpha}^2$ or (2.25c) for the frequency scales of interest. Then (3.7) and (3.9) reduce to

$$\begin{aligned} R_{W}^{[b]}(\tau) &= e^{-\beta\tau N} \bigg(\cos \delta\tau N - \frac{\beta}{\delta} \sin \delta\tau N \bigg), \\ \overline{\zeta}^{2}(t, \gamma = 0)^{[b]} &= 2 \bigg[1 - e^{-\beta t N} \bigg(\cos \delta t N + \frac{\beta}{\delta} \sin \delta t N \bigg) \bigg], \\ \overline{\zeta}^{2}(\infty, \gamma = 0)^{[b]} &= 2. \end{aligned}$$
(3.10)

The predicted forms of $R_W(\tau)$, the Lagrangian autocorrelation function, are plotted in figure 5 for the two different spectral forms used in (3.6) and (3.7). The differences are not marked; both display the negative values when $N\tau > 1$, which have been remarked on before by others (e.g. Csanady 1964; Pasquill 1974), a feature not found in calculated or observed forms of R_W in neutral conditions. For the computations shown in figure 5, we have taken the wave-damping factor $\beta(=k/N) = 0.8$ to be close to the value $\sqrt{\frac{1}{2}}$ suggested by Cerasoli's (1978) observation. In this case $\delta = 0.6$.

In figures 6(a-d) we plot various aspects of the development of the 'plume depth' $(\overline{Z^2}(t))^{\frac{1}{2}}$; it is most economical to discuss all of them together with the analytical results just obtained. First, it is clear from the forms of $\tilde{\Phi}(\omega)$, the Lagrangian pressure-gradient spectra, and the transfer functions $\tilde{P}^{-1}(\omega)$ and $\omega^2 \tilde{P}^{-1}(\omega)$ (for the limits $Nt \to \infty$ and $\gamma = 0$) as defined in (2.19) and sketched in figure 3, that to the first approximation

$$\int_{-\infty}^{\infty} \tilde{P}^{-1} \tilde{\Phi}(\omega) \, d\omega \sim \tilde{\Phi}(\omega=1) \beta^{-1}, \quad \int \omega^2 \tilde{P}^{-1} \tilde{\Phi}(\omega) \, d\omega \sim \tilde{\Phi}(\omega=1) \beta^{-1}, \quad (3.11)$$

so that $\overline{\zeta^2}(\infty, \gamma = 0) \sim 1$. However, figures 6(c, d) show that as α decreases (i.e. increasing stratification) $\overline{\zeta^2}(\infty, \gamma = 0)$ increases, but tends to a limit in the case of the spectrum [a], and increases continuously in the case of [b]. The reason is that $\int \Phi \tilde{P}^{-1} d\omega$ is a constant fraction of $\int \omega^2 \tilde{P}^{-1} \Phi d\omega$ for [a] when $\alpha \leq 1$, but a steadily increasing proportion of $\int \omega^2 \tilde{P}^{-1} \Phi d\omega$ for [b], because $\Phi^{[b]}(\omega)$ is decreasing when $\alpha Re \leq 1$.

Secondly, figure 6 shows that the normalized asymptotic plume width $\overline{\zeta}^2(t)$ is rather insensitive to the form of $\tilde{\Phi}(\omega)$, when $tN \ge 1$, $\gamma \ll 1$, and $tN\gamma^2 \ll 1$. This is because $\overline{Z^2}(\infty)$ and $\overline{W^2}$ are, to first order, both proportional to $\tilde{\Phi}(\omega = N)$. This generalization is most likely to be true when N lies within the range of frequencies of the inertial subrange. (One might expect significantly different results between low-Reynoldsnumber laboratory turbulence (when there is a very limited inertial subrange) and field observations.) When $Nt \gtrsim \gamma^{-2}$, the effect of fluid elements changing their density appreciably increases $\overline{Z^2}(t)$ for the spectrum [b], but not for the spectrum [a], because the growth of $\overline{Z^2}(t)$ depends crucially on the form of $\tilde{\Phi}(\omega)$ as $\omega \to 0$, as (3.3) shows. In neutral flows, $\tilde{\Phi}(\omega = 0) = 0$, if $\ddot{Z} = H(t)$ and $\ddot{Z^2}$ is bounded (Lin 1960). Whether or not there can be some small but finite value of $\tilde{\Phi}(\omega)$ as $\omega \to 0$ in uniform stable flows is an important but unanswered question. At such scales other effects such as inhomogeneity of the turbulence may make the question irrelevant.

4. The density flux, motions of fluid elements and molecules, and some dynamical consequences

4.1. Density flux

The average density flux F_{ρ} is defined in Eulerian terms, denoted by $\overline{(\)}^{(\mathbf{E})}$, as the time or ensemble average of the product of the local density and vertical velocity at (\mathbf{x}, t) : $F_{\rho} = \overline{(\rho + \rho')(\mathbf{x}, t)w(\mathbf{x}, t)}^{(\mathbf{E})} = \overline{\rho'(\mathbf{x}, t)w(\mathbf{x}, t)}^{(\mathbf{E})}.$ (4.1)

In a homogeneous stationary flow F_{ρ} is uniform and can be expressed in Lagrangian terms as the ensemble (or time) average for one, or all, fluid elements of the product of the density $(\rho + \rho')(t)$ and vertical velocity W(t). Therefore

$$F_{\rho} = \overline{(\rho + \rho')(t) W(t)}. \tag{4.2}$$

Before calculating F_{ρ} with our model, it is instructive to consider the causes of density fluctuations and density flux in a stably stratified flow.



FIGURES 6(a-c). For caption see facing page.

232



FIGURE 6. (a) Forms of solution for $\overline{Z^2}(t)$ for strong stratification (curve $\bigoplus \alpha (= 1/NT_H) \leq 1$) and weak stratification (curve $\bigoplus, \alpha \geq 1$) and very weak effects of density mixing between fluid elements $(\gamma \ll 1)$, showing also the effects of different spectral forms on the asymptotic behaviour. (b) Development of the square of the plume depth, $\overline{\zeta^2}(t)$ for two forms of spectra [a] and [b] from (3.6), (3.7). ---, Asymptotic values as $t \to \infty$ ($\alpha = 1, \gamma = 0; \beta = 0.8, \hat{\alpha} \geq 1$). (c) Asymptotic plume depth squared $\overline{\zeta^2}(\infty)$, when $\gamma = 0$, as a function of stratification for two spectra of the form [b], that is $\Phi(\omega = 0) \neq 0$. $\Box, \Phi(\omega, \alpha) = \exp(-|\omega|/\hat{\alpha}); -- \Phi -, \Phi(\omega, \alpha) = 1/(\omega^2 + \hat{\alpha}^2)$. (d) Asymptotic plume depth squared for varying values of stratification and of diffusity γ , when $\Phi = \omega^2/(\omega^2 + \alpha^2)$ (type [a] spectrum). -- $\Phi -, \gamma = 0.0; \Box, 0.2; \blacksquare, 0.5; \blacktriangle, 1.0$.

Consider the expression for $\rho'(t)$ obtained by integrating (2.2):

$$\rho'(t) = \int_0^t \left(\mathscr{G} W(t') + \frac{d(\rho + \rho')(t')}{dt'} \right) dt'.$$
(4.3)

This (exact) expression shows how density fluctuations are caused by the vertical advection of the fluid elements with different densities and by the actual change in density of fluid elements produced by mixing with neighbouring elements.

Multiplying (4.3) by W(t) and evaluating the integral gives[†]

$$F_{\rho} = \frac{1}{2} \frac{d\overline{Z}^2}{dt}(t) \mathscr{G} + \overline{\Delta\rho(t) W(t)}.$$
(4.4)

The first, advective, term was given by Corrsin (1951). The second 'mixing' term involves $\Delta \rho(t)$, the change in density of a fluid element, which is equal to

$$\int_0^t (d(\rho+\rho')/dt')\,dt'$$

Expressed in non-dimensional form, (4.4) becomes, using (1.1) and (1.2),

$$\mathscr{F} \equiv \frac{gF_{\rho}}{\rho_0 \ \overline{W^2}N} = \frac{K_{\rho}}{\overline{W^2}/N} = \mathscr{T} + \frac{g}{\rho_0 \ \overline{W^2}N} \ \overline{\Delta\rho(t) \ W(t)}, \tag{4.5}$$

where $\mathscr{T} = NT_{\rm L}$, $T_{\rm L}$ being the Lagrangian velocity integral timescale, and $K_{\rho} = F_{\rho}/\mathscr{G}$, K_{ρ} being the the eddy diffusivity of density. Equation (4.3) shows how

 $[\]dagger F_{\rho}$ is here the flux produced by fluid elements at time t which were released at time t = 0. Since F_{ρ} is found to be independent of t, (4.4) is valid for all time.



FIGURE 7. Schematic diagram showing net vertical density transfer between fluid elements having restricted vertical motions (after Puttock 1976).

a vertical flux can exist even if $T_{\rm L} = 0$, and if the mean-square vertical displacement of fluid elements is bounded. The process by which the fluid elements transfer density while performing bounded oscillations is illustrated in figure 7, and is discussed in greater detail in Puttock (1976).

The dimensionless density flux \mathscr{F} can be calculated with our Lagrangian model. From (4.2), (4.5), (2.12) and (2.13),

$$\mathscr{F} = \int_{-\infty}^{\infty} (\gamma + i\omega) \,\tilde{\Phi}(\omega) \tilde{P}^{-1}(\omega) \,d\omega, \qquad (4.6)$$

whence using the Hermitian symmetry of $\tilde{\Phi}(\omega)$ and (3.3)

$$\mathscr{F} = \gamma \int_{-\infty}^{\infty} \tilde{\Phi}(\omega) \tilde{P}^{-1}(\omega) \, d\omega = \frac{1}{2} \gamma \overline{\zeta^2}(\infty, \gamma = 0). \tag{4.7}$$

Taking $\overline{\zeta}^2(\infty, \gamma = 0)$ as given by (3.8) and (3.9) for the two models of pressure-gradient spectra defined in (2.25*a*, *b*),

$$\mathscr{F}^{[a]} = \frac{\gamma}{1 + 2\alpha\beta}, \quad \mathscr{F}^{[b]} = \gamma \left(1 + \frac{2\beta}{\alpha R e^{\frac{1}{2}}}\right). \tag{4.8}$$

Thus, in general, \mathscr{F} does not have the same value for both spectra. (This would only occur in a flow where N lies in the inertial-subrange part of the pressure-gradient $(\Phi(s))$ spectrum, in which case the inertial range would not have the form of (2.24).)

Comparing the magnitudes of the terms \mathscr{T} and \mathscr{F} in (4.5) gives a measure of the relative contribution to the density flux by the rate of increase of the mean-square displacement of marked fluid elements. From (2.22*a*),

$$\mathscr{T} = \frac{\pi \gamma^2 \Phi(0)}{(1+2\beta\gamma)^2}.$$
(4.9*a*)

For spectrum [a] $\mathscr{T}^{[a]} = 0$, but for spectrum [b] when $\alpha \ge 1$, $\gamma \ll 1$, using (2.25c),

$$\mathcal{F}^{[b]} = 2\beta\gamma^2. \tag{4.9b}$$

Thus, when $\gamma \ll 1$, $\overline{\Delta\rho(t) W(t)}$ is $O(\gamma^{-1})$ larger than $\mathscr{G} d\overline{Z^2}/dt$. Another way of looking at this result is to compare neutral flows, where

$$\frac{d\overline{Z^2}}{dt} = 2K_{\rho} \quad \text{when} \quad \frac{t}{T_{\rm L}} \gg 1, \tag{4.10a}$$

with stably stratified flows, where

$$\frac{d\overline{Z^2}}{dt} = 2\overline{w^2}T_{\rm L} = O\left(\frac{NK_{\rho}^2}{\overline{W^2}}\right) \ll K_{\rho}. \tag{4.10b}$$

In neutral flows the eddy diffusivity is proportional to the rate of growth of Z^2 , because the flux is carried by the marked fluid elements moving across the *whole flow*. In stable flows the flux is carried by limited fluid-element displacements and mixing between the elements. The flux produced by the large-scale displacements ($\propto d\overline{Z^2}/dt$) is only a very small proportion of the total flux.

So far we have only considered $\gamma \ll 1$. What about larger values of γ (corresponding to weaker stabilities)? From the contour integrals the large- αRe limit (high-Reynolds-number turbulence) for $R_W(t)$ with spectrum [b] can be derived:

$$R_{W}^{[b]}(\tau) = e^{-\eta\tau} \bigg[\cos \Lambda t - \frac{\eta (\Lambda^2 + \eta^2 - \gamma^2)}{\Lambda (\Lambda^2 + \eta^2 + \gamma^2)} \sin \Lambda t \bigg], \tag{4.11}$$

where

$$\eta = \beta + \frac{1}{2}\gamma, \quad \Lambda = (1 + \beta\gamma - \beta^2 - \frac{1}{4}\gamma^2)^{\frac{1}{2}}.$$

Figure 8(a) shows the non-dimensional vertical density flux \mathscr{F} as a function of γ and α for spectrum [a], evaluated from (4.7) and (4.4). Figure 8(b) shows the density flux and the non-dimensional integral timescale (which indicates the flux of marked fluid elements) for spectrum [b] with $\alpha Re^{\frac{1}{2}} \ge 1$. Note how \mathscr{F} (= $NT_{\rm L}$) becomes of the same order of magnitude as \mathscr{F} for $\gamma = O(1)$, but is negligible for $\gamma \ll 1$. When $\gamma \gtrsim O(1)$, the density flux is mainly carried by fluid elements rather than being transferred between them.

4.2. Diffusion problems

Mixing between fluid elements also affects the diffusion of marked molecules or particles, e.g. dye, gas or smoke, released into the flow at the source (figure 1). The result (3.3b) suggests that, when this mixing is slow compared with the turbulent motions' timescale ($\gamma \ll 1$), a 'plume's' growth may level out with a depth of order $(\overline{W^2})^{\frac{1}{2}}/N$, and then, after a time $N^{-1}\gamma^{-2}$, may grow parabolically like $t^{\frac{1}{2}}$. Since the latter growth rate is only possible if the molecules of the ambient fluid diffuse into and out of the fluid elements, the marked molecules or particles must also diffuse out of the original marked fluid elements which passed through the source.

Since, as we have demonstrated, the mixing between fluid elements can be more rapid than the rate of growth of the fluid elements' displacements, this means that the mean square displacement of the marked molecules $(\sigma_Z^{(p)})^2$ may grow faster than that of the fluid elements. In that case

 $\sigma^{(p)2}(t) > \overline{Z^2}(t)$

where

$$\sigma_{Z}^{(p)2}(t) = \frac{\int \int Z^{2}\overline{C}(\mathbf{x}, t) d\mathbf{x}}{\int \int \int \overline{C}(\mathbf{x}, t) d\mathbf{x}},$$
(4.12)

and \overline{C} is the mean concentration of marked molecules at time t.



FIGURE 8(a). For caption see facing page.

Some contaminants may take the form of small particles much larger than molecules, but much smaller than the smallest, Kolmogorov, scales of turbulent motion. Such particles follow the motion of the *fluid elements* into which they are released (Csanady 1973, chap. 2). Since in stable flows marked molecules may diffuse more rapidly than marked fluid elements, it follows that, unlike the case of neutral or unstable turbulent flows, there may be measurable differences in the ultimate growth rates of 'plumes' of particles, such as smoke or liquid droplets, and of 'plumes' of molecules with the same scale as that of the fluid (e.g. dye or gas).

4.3. Dynamical consequences of mixing

The rate of increase of potential energy per unit height in a homogeneous, inviscid, non-diffusive incompressible flow in a region confined by solid boundaries, in which there is a mean density gradient, can be shown (e.g. by using the energy equation of Milne-Thompson 1968, p. 83) to be

$$\frac{dV}{dt} = gF_{\rho}$$

(see also Pearson 1981, pp. 40, 41).

Thence, from the expression for F_{ρ} in (4.5), if $T_{\rm L} > 0$ and if fluid elements do not mix, the potential energy would increase without limit. Therefore for a turbulent flow



FIGURE 8. (a) Non-dimensional vertical density flux \mathscr{F} for $\mathbf{\Phi} = \omega^2/(\omega^2 + \alpha^2)$ – spectrum [a] (note $\mathscr{T} = 0$). \Box , $\alpha = 0.5$; ×, 1.0; \triangle , 2.5; +, 10.0. (b) Non-dimensional flux \mathscr{F} and integral timescale \mathscr{T} for $\mathbf{\Phi} = 1/(\omega^2 + \alpha^2)$; $\hat{\alpha} = 200$ – spectrum [b]. \blacksquare , \mathscr{F} ; \blacktriangledown , \mathscr{T} . The arrows indicate the observed range of \mathscr{F} in the atmosphere (Hunt 1982; Hunt *et al.* 1982).

started from rest (e.g. by a grid) with a finite amount of kinetic and potential energy of order $\rho \overline{w^2}$, it follows that $\overline{Z^2} \sim \overline{w^2}/N^2$.

But suppose there is an energy supply (e.g. by a shear flow), in a dissipative but not diffusive flow, then is it possible that $d\overline{Z^2}/dt > 0$? If this were possible then the buoyancy forces would amplify $\overline{H^2}(t)$ and thence $\overline{w^2}(t)$. Therefore it would be impossible for w(t) and H(t) to be stationary processes. Thus if the turbulence is stationary, an energy supply can only lead to more viscous dissipation. In other words, if $d\overline{Z^2}/dt > 0$ and $\gamma = 0$, then the velocity and acceleration cannot be stationary random functions, because of the dynamical effect of particle displacements in a stratified flow.

So *if* energy is supplied and *if* the velocity and acceleration are s.r. functions, then all the energy *must* go into viscous dissipation. In most real, steady, stably stratified turbulent flows, where $\gamma \neq 0$, most of the shear energy (typically $\frac{4}{5}$) goes into viscous dissipation and a small part (typically $\frac{1}{5}$) is transferred into potential energy because of mixing between fluid elements (Turner 1973, §5.2). We are now in a position to answer the question of when molecular diffusion and mixing of density between elements can be ignored in the *initial stage* of plume growth. Substituting (4.3) into the inviscid momentum equation for a fluid element, multiplying by W(t) and averaging over our ensemble of marked elements, gives

$$\frac{1}{2}\frac{d}{dt}\overline{Z^2} + N^2\frac{d\overline{Z^2}}{dt} = -\left(\frac{\dot{Z}}{\rho_0}\frac{\partial p'}{\partial z}\right) - \frac{g}{\rho_0}\overline{\Delta\rho}\,\overline{W}(t). \tag{4.13}$$

The density-flux term $\overline{\Delta\rho W}(t)$ in (4.13) for an ensemble of marked elements from a source is less than the second term in (4.4) until a long time after the marking of the particles. Since in strong stratification, for $t \sim N^{-1}$,

$$N^{2} \frac{d\overline{Z}^{2}}{dt} \sim N^{2} \left(\frac{\overline{Z}^{2}}{N^{-1}}\right) \sim N\overline{w}^{2},$$
$$\frac{g\overline{\Delta\rho} \,\overline{W}(t)}{\rho_{0}} \lesssim \frac{gF_{\rho}}{\rho_{0}},$$
(4.14)

and since, from (4.4),

the condition for the neglect of density in (4.13) in estimating $\overline{Z^2}(t)$ is

$$F_{\rho} \ll \frac{\overline{W^2} N \rho_0}{g}. \tag{4.15}$$

From (4.5) and (4.8), F_{ρ} may be estimated as

$$F_{\rho} \sim \frac{\gamma \mathscr{G} \overline{W^2}}{N} \sim \frac{\gamma \rho_0 N \overline{W^2}}{g}.$$
(4.16)

So the condition (4.15) for the neglect of mixing between fluid elements in calculating $\overline{Z^2}$ reduces to $\gamma \ll 1$.

5. Comparison with experiment

The theory can be compared with observation and measurements of diffusion from point sources and density fluctuations in the atmosphere, in wind-tunnel boundary layers and in grid-generated turbulence. An example of each of the first two types is given, as well as a discussion of the observations of BHMS. For brevity, we shall use the term 'plume' depth and the symbol σ_z to denote $(\overline{Z^2})^{\frac{1}{2}}$, the r.m.s. particle displacement or the root of the second moment of mean concentration distribution.

5.1. Boundary-layer results

Hilst & Simpson (1958) report experiments on the vertical diffusion of a passive tracer released from a point 56 m above ground in a stably stratified atmosphere. The variance σ_z^2 of vertical distribution of the tracer was measured at each of four distances from the point of release, over five sets of meteorological conditions (see table 1). It is revealing to replot their data for variance against diffusion time in a non-dimensional form. Time is non-dimensionalized with N, the buoyancy frequency, and variance with $N^2 U^{-2}$, where U is the mean horizontal velocity measured at approximately the source height. It would be preferable to non-dimensionalize with $\overline{w^2}$, the vertical velocity variance, but this flow statistic was not reported. The resulting composite graph for their five runs is shown in figure 9.

Now (3.1) may be rewritten in non-dimensional form

$$\frac{\overline{Z^2}(t)}{U^2 t^2} \approx \frac{w^2}{U^2},\tag{5.1}$$

Experiment number	U (m s ⁻¹)	$N \atop (s^{-1})$	$\overline{W^2}/U^2 \times 10^3$
A	6.3	0.025	1-06
В	5.1	0.023	1.02
C	5.2	0.015	1.29
D	4.6	0.009	1.27
E	6.0	0.034	0.74

 $\overline{W^2}/U^2$ is estimated from (5.1) and hence may be an underestimate by up to a factor of 2

TABLE 1. Hilst & Simpson's (1958) atmospheric diffusion experiments



FIGURE 9. Hilst & Simpson (1958) field data on plume widths replotted in non-dimensional form (see table 1): \diamondsuit , test A; $\textcircled{\bullet}$, test B; \bigcirc , test C; \bigstar , test D; \blacksquare , test E. Theoretical results from (3.3*a*): ---, $\gamma \leq 0.1$, $\overline{\zeta^2}(\infty, \gamma = 0) = 2$; ----, $\gamma \approx 0.4$; ($\mathcal{F} = 0.08$), $\overline{\zeta^2}(\infty, \gamma = 0) = 2$.

indicating that all the dispersion curves should coincide for small time. Applying this formula to the first data point of each set provides an estimate for $\overline{w^2}U^{-2}$ (see table 1). (In discussing these experiments we assume $\sigma_z^2 = \overline{Z^2}$.)

The values taken for N were estimated by assuming a uniform temperature gradient between 30.5 m and 91.5 m, the heights at which the temperatures were measured.

Figure 9 then shows that the rate of growth of plumes in stable conditions is quite suddenly reduced or stopped when $Nt \sim 1$.

It is striking that the data points from tests A, B and C (table 1) lie close to the same curve and interesting that the two exceptions to this, tests D and E, are at either extreme of the range of atmospheric stabilities occurring during these experiments. Test D had the least-stable conditions ($N = 0.009 \text{ s}^{-1}$) and the non-dimensionalized dispersion results lie below the general level.

For the most stable test E (corresponding to the smallest value of $\overline{w^2}/U^2$ in table 1), where a final plume depth is unambiguously established, its non-dimensional depth σ_z is close to that predicted by (3.8) or (3.9) in the appropriate range of NT (see figure 9), and $\gamma \leq 0.1$.



FIGURE 10. Picture of smoke plume from the chimneys of the Kyndby power station drifting 20 km past Risø towards Roskilde. Picture taken at Risø (Denmark) (Koføed-Hansen 1962).



FIGURE 11. Vertical spread from a continuous source placed in a turbulent stratified wind tunnel boundary layer. For details see text and Chaudhry & Meroney (1969). \bigcirc , 10% of C_{mx} ; \times , 50% of C_{mx} ; \odot , 100% of C_{mx} (the maximum concentration given at x).

The results of tests A, B, C are also consistent with the theory, if some mixing between fluid elements is taking place. The theoretical curve (3.3) is plotted with $\mathscr{T} = 0.08$, a value that would be observed if $\mathscr{F} \approx 0.2$ and $\gamma \approx 0.4$, according to figure 8. These variations in γ are consistent with measured variations in \mathscr{F} ; but the cause of this variation is unknown. This curve is similar to the data of Högstrom (1964).

Another documented example of a plume is that of Koføed-Hansen (1962) at Risø, where a plume at 150 m above the ground in an 8 m s⁻¹ wind, was observed to travel for 20 km in stable conditions ($N \approx 3 \times 10^{-2} \, \text{s}^{-1}$), with little vertical spread ($\sigma_z \approx 7 \, \text{m}$) (figure 10). In this case some turbulence measurements at 56 m indicated $(w^2)^{\frac{1}{2}}/U \approx 0.02$ (the turbulence was quite intermittent), so that $(2w^2)^{\frac{1}{2}}/N \approx 7 \, \text{m} - q$ uite a reasonable estimate for the plume depth which is consistent with a value of $\gamma = 0.1$. Other examples may be found where this estimate is found to be useful. It is probably not appropriate where the source is within a distance of σ_w/N of the surface.

Chaudhry & Meroney (1969) made measurements of the dispersion of a passive tracer, from a point source, in a large thermally stratified wind tunnel. Their elevated source 0.2 m (8 in.) above the tunnel floor is in the upper part of the boundary layer and so the effect of shear on vertical diffusion was small, certainly for the first few seconds. Their results for concentration distribution downstream of the elevated source (see figure 11, which is figure 35 in their report), show signs of an abrupt reduction of spreading of the 50 % of maximum concentration contour, on the side away from the floor, about one second after leaving the source. At z = 0.20 m, the mean horizontal velocity is about 2.4 m s⁻¹ (8 ft s⁻¹). At 2.44 m downwind of the source $Nt \simeq 1.5$.

From the measurements of the intensity of vertical velocity fluctuations (Arya 1968), under these conditions in the same tunnel, we find

$$(2w^2)^{\frac{1}{2}}/N \approx 0.08 \text{ m.}$$
 (5.2)

To compare the theoretical estimate (5.2) for the plume depth σ_z with figure 11, it is assumed that the vertical concentration profile is approximately Gaussian. Thence figure 11 shows that $\sigma_z \approx 0.064$ m (since the 50 % contour is $1.2\sigma_z$).

5.2. Grid-turbulence results

Grid-generated turbulence decays away from the grid and the development and decay of velocity fluctuations is associated with the growth and final decay of density perturbations. This must be borne in mind when interpreting the results of such diffusion experiments.

In an earlier paper (BHMS) measurements are presented of the development of a plume of passive tracer in stably stratified grid turbulence, for a range of mean density gradients. Velocity fluctuations were also measured in two cases: neutral flow and for stable flow with U/MN = 1.06, where M is the grid mesh size. They find that horizontal plume growth, σ_y , is weakly if at all influenced by the density stratification even for $U/MN \approx 1.06$. The growth of the plume in the vertical, σ_z , is attenuated by the density stratification, with σ_z increasing to a maximum and then either a slight decrease to a constant plume width, or no decrease and a maintained maximum plume width (see BHMS, figures 3, 4b). The final vertical plume width ($\sigma_z(\infty)$) is a function of the Froude number U/(MN). In their figure 8, BHMS plot the dependent variable $\sigma_z(\infty) N/w'_s$ where w'_s is the r.m.s. vertical velocity at the source, against U/(MN).

Their results are in general agreement with the theory of this paper for nondimensional plume depth, with $(\sigma_z(\infty) N/w'_s)^2$ corresponding to $\overline{\zeta}^2(\infty, \gamma = 0)$ (see §3.2, (3.3)), of order unity. However, as already noted there are two main ways in which the experiments differ from the theory for stationary turbulence: (i) the turbulence velocities decay downstream of the grid, and (ii) the range of densities of fluid marked by the tracer depends on the position of the source (relative to the grid) and the Froude number.

In neutral flows the direct effects of velocity decay on diffusion are balanced by increases in the length- and timescales of the turbulence (Batchelor & Townsend 1956). So, at least until the final stage of decay, the plume width increases ($\sigma_z \propto t^{\frac{1}{2}}$) just as in the stationary case. However, in a stably stratified fluid this increase in Lagrangian length- and timescales for the vertical velocity is inhibited by buoyancy forces. That is to say the vertical migration of fluid elements is severely restricted by the density stratification and so the extent of diffusion cannot grow as the turbulence decays, as it does in the neutral case.

For the case U/(MN) = 1.06 the BHMS results (figures 3, 5b) show that the plume has reached its maximum vertical width by 15 mesh lengths downstream, while the vertical velocity fluctuations are not decaying any faster than in the neutral case. So, as BHMS conclude, the reduction in plume growth is not due to velocity fluctuations decaying more rapidly as the Froude number decreases.

Lange (1974) gives explicit measurements of density variances $\overline{\rho'^2}$ behind a grid in a linear density gradient. His result shows that $\overline{\rho'^2}$ reaches a maximum at time $Nt \sim 1$ behind the grid and then decays to zero, but more slowly than the velocity fluctuations (as would be expected). He argues that, if molecular diffusion can be neglected, then $\overline{\rho'^2}$ is a measure of the variance of fluid particle displacements from their original level. Details comparisons between his results and the theory of this paper are given in Pearson (1981). Atmospheric measurements of $\overline{\rho'^2}$ in stable conditions are presented and discussed by Hunt *et al.* (1982) and Hunt (1982).

In the BHMS experiments, the source was at x/M = 4.7 downstream of the grid. Lange's results suggest that it may be sampling the full range of densities possible



FIGURE 12. Theoretical predictions $\Phi = \omega^2/(\omega^2 + \alpha^2) - \text{case } [a] (\gamma = 0)$. No decay, from (3.6), with $\alpha = 2$. Decay, from (5.3), with $\alpha = 2$, $Nt_0 = 1.5$.

(i.e. outside the region of growth of density perturbations) for $U/(MN) \leq 4.7$. In some senses the final plume width is a measure of the range of densities sampled at the source.

A crude attempt to model the kinematical effects of decaying turbulence is made as follows. (a) Assume that decay of turbulence can be decoupled from other processes so that on a 'local' timescale we can use vertical velocity correlations calculated for the stationary case. (b) Assume that these 'local' correlations are stationary in time. Explicitly we assume that the vertical velocity following a fluid element is the product of a deterministic decaying amplitude and a stochastic signal. The decay function is given by the experimental form and the random signal has known autocorrelation function. Vertical diffusion is then calculated from the exact Taylor (1921) relation

$$\sigma_z^2(t) = \int_0^t \int_0^t \overline{W(t^*) \ W(t')} \ dt^* \ dt'.$$
(5.3)

This has been evaluated numerically, by standard routines on an IBM 370/165, for the velocity autocorrelation of (3.6) corresponding to pressure-gradient spectrum (2.25*a*). Typical results show little qualitative change from the equivalent stationary turbulence results. A comparison is given in figure 12. After reaching a maximum at $Nt \approx 3$, the plume depths decline somewhat. This decline is mainly due to the explicit decoupling in the model of turbulence decay from the other dynamics of the problem. The data is ambiguous as to whether such a decline is real. As the further behaviour of the plume after reaching a maximum is probably not well modelled, we choose the maximum computed value of σ_z , $\sigma_{z \max}$, for comparison with data.

The non-dimensional frequency scale $\alpha = (NT)^{-1}$ of the pressure-gradient spectrum is assumed to be proportional to the Froude number

$$\alpha = c \frac{U}{MN} \tag{5.4}$$

because the decay of the grid turbulence shows that the turbulent timescale given by the grid timescale $c^{-1}M$

$$T = \frac{c^{-1}M}{U}.$$

Figure 13 shows a comparison of three types of normalized plume depths as a function of the Froude number: (a) the experimental data of BHMS, (b) calculated value of



FIGURE 13. Asymptotic plume depths. \bigcirc , BHMS data. Computations with spectrum [a] and two different values of α : —, no decay – see (3.8); ---, with decay (see text). (i) $\alpha = U/NM$, (ii) $\alpha = \frac{1}{3}U/NM$.

 $\sigma_{z\max} N/w'_{s}$ from (5.3) (see Pearson 1980 for details); and (c) equivalent results for the final 'plume' depth in stationary turbulence from (3.8). The spectrum

$$\tilde{\Phi}^{[a]} \propto \frac{\omega^2}{\omega^2 + \alpha^2} \tag{5.5}$$

was chosen for comparison as (3.10) shows the same trend with α or Froude number as the BHMS data. (b) and (c) curves are shown for two plausible values of $c = \alpha MN/U$. Two points are clear. (i) The effects of modelling any turbulent velocity decay are small at low Froude number. This is because the final plume depth is reached in a time short compared with that of turbulence decay for large N. (ii) The experimental results (certainly at low Froude number) are consistently greater than those of the simple theory. With the normalization used $(\sigma_z(\infty) N/w'_s)$ the experimental values exceed the theoretical maximum (from (3.8)) of $\sqrt{2}$, for $U/(MN) \leq 1$. This suggests that w'_s is not the most appropriate velocity for normalization. Lange's results show that the density-fluctuation field is set up by a time $Nt \approx 1$, and then only weakly decays until about $Nt \approx 6$ at lower Froude numbers (large N). Therefore, the density field is generated at an earlier time when turbulent velocity intensities are larger, so that a wider range of densities than suggested by w'_s is sampled at the source. (This means that $\overline{Z}^{2(q)}$ would be a lower proportion of \overline{Z}^2 than is indicated by (3.5).)

5.3. Lagrangian autocorrelation measurements

Frenzen (1963) directly measured Lagrangian velocities in stratified grid turbulence by following neutrally buoyant particles photographically. He computed vertical

Vertical diffusion in stratified turbulent flow 245

velocity autocorrelations and found them qualitatively to be of the form computed from the theory of §3 and shown in figure 5. Unfortunately, he applied the Batchelor (1952) transformation to his data in an attempt to correct for turbulence decay. Lange (1974) pointed out in discussing Frenzen's results that the transformation cannot be valid in this case, where the autocorrelation function has a negative loop and where the 'local' integral timescale $T_{\rm L}(t)$ defined by

$$\frac{1}{2}\frac{dZ^2}{dt} = \overline{W^2}(t) T_{\rm L}(t)$$
(5.5)

is approximately zero. Frenzen's results cannot therefore be quantitatively correct. However, they, like the plume measurements of BHMS, are in agreement with the general predictions of our theory.

6. Conclusions

In this paper we have developed a Lagrangian statistical model, albeit rather speculative, for vertical diffusion in stably stratified turbulent flows. The dimensionless form $\overline{Z^2}N^2/\overline{w^2}$ relating vertical displacement and vertical velocity statistics with the ambient stratification is identified. It is found that experimental results on diffusion from point sources support the use of this ratio. The model shows how buoyancy forces can restrict the vertical motions of finite fluid elements or control volumes ((i) in §1). A rough equipartition then exists between turbulent kinetic energy and the potential energy of the randomly varying density field.

Growth of the plume from a point source is sharply reduced after a time of order N^{-1} . This is a feature not reproduced if a gradient transport model with associated eddy diffusivity is used ((ii) in §1). A vertical flux of density through the fluid and a continued vertical growth of plume occur through the exchange of density between fluid elements by small-scale processes.

Stable stratification has the effect of introducing a negative loop into the Lagrangian autocorrelation function, $R_W(t)$, of the vertical velocity. This negative loop allows the integral timescale T_L to assume a very small or zero value, consistent with the restriction of vertical diffusion.

The model focuses attention on the dynamic consequences of density fluctuations and their connection with vertical displacements (Lange 1974; Frenzen 1964). This provides a framework for understanding, at least qualitatively, the complex interaction of density and velocity fields behind a grid in a stable medium. The experiments of BHMS provide some general support for the model but introduce further difficulties because of their inherent non-stationarity.

It is hoped that both the specific, and more general, results of this theory will be tested experimentally and in computer simulations by Lagrangian measurements or computations of Z, ρ' , and $\partial p/\partial z$.

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Appendix. Previous random-force models

Lin & Reid (1963) consider the motion of fluid particles satisfying the equation (in our notation) dW(t)

$$\frac{dW(t)}{dt} = -fW(t) + H(t), \tag{A 1}$$

where f represents viscous damping and H a stationary random pressure-gradient force. This is analogous to the Langevin equation for the Brownian motion of suspended particles (Uhlenbeck & Ornstein 1930). Their theory requires that H(t) is only correlated over a timescale short compared with that of observations – i.e. Hhas a 'white-noise' or flat frequency spectrum. They recover Taylor's (1921) results for dispersion at small and large times. Krasnoff & Peskin (1971) (see also Krasnoff 1970) developed Lin's model further. They assumed that the random force had a finite timescale τ_H (note: not of the same magnitude as T in this paper). By restricting attention to isotropic turbulence they derived an energy equation from (A 1). Stationarity in time gives an exact balance between viscous dissipation and the average rate of energy input to a fluid particle by the pressure gradient

$$\overline{W(t) H(t)} = f \overline{W^2}.$$
 (A 2)

Further use of the stationarity condition suggests the choice

$$R_H(\tau) = \exp\left[\frac{(1-\alpha^*)/|\tau|}{\tau_H}\right]$$
(A 3)

for the autocorrelation coefficient of the random force. Here

$$\alpha^* = f \tau_H, \tag{A 4}$$

and as f^{-1} is a characteristic timescale of the particle velocity, α^* is expected to be a very small number in large-Reynolds-number turbulence. The frequency-power spectrum corresponding to (A 3) for the random pressure gradient is, to within a multiplicative constant,

$$\Phi^*(s) \propto \frac{c\tau_H^{-2}}{s^2 + (1 - \alpha^*)^2 / \tau_H^2}.$$
 (A 5)

Introducing λ , the Taylor microscale of the turbulence (so that the dissipation rate $\epsilon = 15\overline{\nu w^2}/\lambda^2$) and the turbulence Reynolds numbers R_{λ} , Re where

$$R_{\lambda} = \frac{(\overline{w^2})^{\frac{1}{2}/\lambda}}{\nu} \equiv (15Re)^{\frac{1}{2}}, \tag{A 6}$$
$$Re = \frac{(\overline{w^2})^{\frac{1}{2}}L_x^{(w)}}{\nu}, \quad L_x^{(w)} = \frac{\epsilon}{(\overline{w^2})^{\frac{3}{2}}},$$

their results can be summarized as follows

$$\begin{split} f &= R_{\lambda}^{-1} \left(\frac{5\epsilon}{3\nu}\right)^{\frac{1}{2}}, \\ \tau_H &= \frac{1}{3} (1 + C R_{\lambda}^{-1}) \left(\frac{\nu}{\epsilon}\right)^{\frac{1}{2}}, \quad C = 0.43 \end{split}$$

 $(\tau_H \text{ is of the order of the Kolmogorov microtimescale})$

$$\alpha^* = CR_{\lambda}^{-1}(1-R_{\lambda}^{-1}).$$

Thus when $Re^{\frac{1}{2}} \gg 1$, this form of $\Phi(s)$ when normalized on $\overline{w^2}$ and N becomes

$$\tilde{\Phi}(\omega, \alpha) \propto [\omega^2 + (\alpha R e^{\frac{1}{2}})^2]^{-1}$$
(A 7)

where $\alpha = NT \sim NL_x^{(w)}/(\overline{w^2})^{\frac{1}{2}}$.

Chadam (1962) attempted to extend Lin's theory to stratified flows, but failed to account satisfactorily for the varying densities and initial velocities of the marked diffusing fluid particles. He did, however, recognize the role of stratification in limiting vertical particle diffusion.

Csanady (1964) modelled the motion of fluid elements that could not change their density in time by the following equations (our notation):

$$\frac{dW}{dt} + \frac{g\rho'(t)}{\rho_0} = H(t), \tag{A 8}$$

$$\frac{d\rho'}{dt} - WG = -D\rho', \tag{A 9}$$

with D a density diffusion constant. So he had no explicit damping term in the momentum equation. He introduces the velocity corresponding to the random acceleration in a neutral flow ct

$$W_1 = \int^t H(t') dt',$$
 (A 10)

and solves (A 8), (A 9) in terms of correlation of the new random variable W_1 . He takes the neutral form $\overline{W_1(t) W(t+\tau)} = \overline{W_1^2} e^{-p|\tau|}.$ (A 11)

$$n_1(t) n(t+1) = n_1 t^2 \cdots t^2$$

Thence the velocity spectrum, and the random 'force' spectrum are respectively

$$\Phi_{W_1} \propto \frac{\epsilon}{s^2 + p^2}, \quad \Phi \propto \frac{s^2 \epsilon}{s^2 + p^2}.$$
 (A 12)

Now, as his formulation (A 8) has no explicit viscous damping, the timescale (p^{-1}) for the spectrum (A 12) is a decay timescale of the velocity fluctuations, of order $L_x^{(w)}/(\overline{w^2})^{\frac{1}{2}}$. (For r.m.s. vertical velocity $(\overline{w^2})^{\frac{1}{2}}$ and lengthscale $L_x^{(w)}, \epsilon$ is such that $\epsilon \sim (\overline{w^2})^{\frac{3}{2}}/L_x^{(w)}$).

The timescale p^{-1} is very much larger than the Krasnoff & Peskin (1971) timescale τ_H , and the two corresponding random-force spectra (A 5) and (2.12) have very different forms. This reflects a different 'level' of modelling. Although not made clear in his paper, we feel that Csanady is really dealing with larger 'fluid elements' than Krasnoff & Peskin, with the viscous processes absorbed into the random force. This is necessary in order to model the effects of buoyancy forces on finite parcels of fluid of approximately the *same* density throughout. Both 'levels' of model are discussed in the paper.

For simplicity, and to ensure that $\overline{W^2} = \overline{w^2}$, Csanady made the specific assumption that D = 0. Because his spectrum (A 12) is such that $\Phi(0) = 0$, he finds an asymptotically constant plume depth. He does not discuss the vertical density flux through the system.

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