J. Fluid Mech. (1980), *vol.* 97, *part* 2, *pp.* 405–416
Printed in Great Britain

Some turbulent diffusion invariants

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(Received 23 October 1978 and in revised form 4 May 1979)

In view of the importance of concentration fluctuations in practical and theoretical problems of turbulent diffusion, there are presented here some invariant properties of the distribution of fluctuations associated with a cloud of contaminant containing a finite quantity Q of material. These properties are invariant provided only that Q is conserved, no assumption whatsoever being made about the random turbulent velocity field. Consequences of the results for (i) steady plumes, (ii) the representation of the distribution of concentration by series of (generalized) Hermite polynomials, and (iii) the relationship of the ensemble mean concentration with the distance-neighbour function, are discussed using experimental evidence.

1. Introduction

A basic problem in turbulent diffusion is the determination of the statistical properties of the concentration field in a cloud of contaminant containing a finite quantity Q of material. It will be supposed throughout this paper that Q remains the same throughout each realization of the dispersion so that, for example, there are no absorbing boundaries or chemical reactions. Naturally it will also be supposed that Q does not vary from realization to realization of the dispersion. But no other restrictions are placed on the contaminant; in particular it does not have to be passive.

Let $\Gamma_0(\mathbf{X}, t)$ be the random distribution of concentration of the contaminant in any realization of the dispersion, where $\mathbf{X}$ is the position vector of a point in space relative to an origin fixed in space (or moving with uniform velocity). Then, by mass conservation,

$$\int \Gamma_0(\mathbf{X}, t)\, dV(\mathbf{X}) = Q, \tag{1.1}$$

where, as throughout this paper, the integration is over all space. Define $\mathbf{X}_c(t)$ and $\mathbf{x}$ by

$$Q\mathbf{X}_c(t) = \int \mathbf{X}\Gamma_0(\mathbf{X}, t)\, dV(\mathbf{X}), \quad \mathbf{x} = \mathbf{X} - \mathbf{X}_c. \tag{1.2}$$

Thus $\mathbf{X}_c(t)$ is the position vector of the centre of mass of the cloud. Its value depends on the distribution of $\Gamma_0(\mathbf{X}, t)$ over the whole cloud and, for fixed t, will vary from realization to realization. Then let $\Gamma(\mathbf{x},t)$ be defined by

$$\Gamma(\mathbf{x},t) = \Gamma_0(\mathbf{x}+\mathbf{X}_c,t) \tag{1.3}$$

(Monin & Yaglom 1975, p. 554). By (1.1) and (1.2)

$$\int \Gamma(\mathbf{x},t)\,dV(\mathbf{x}) = Q, \quad \int \mathbf{x}\Gamma(\mathbf{x},t)\,dV(\mathbf{x}) = \mathbf{0}. \tag{1.4}$$

Thus $\Gamma(\mathbf{x},t)$ is the distribution of concentration in the framework of relative diffusion. There are important conceptual differences between relative and absolute diffusion (the latter requires examination of the statistical properties of $\Gamma_0(\mathbf{X},t)$). Relative diffusion has important advantages over absolute diffusion, both theoretically (Richardson 1926; Batchelor 1952) and practically (Monin & Yaglom 1975, pp. 554, 577).

One fruitful field of turbulence research has been the search for invariants, that is statistical properties of the random velocity or concentration fields that remain constant as time evolves. Such invariants provide a means of checking more complicated calculations or, as in the case of conservation of energy in mechanics, of suggesting alternative methods of modelling the evolution. Furthermore they usually have simple physical interpretations. Two classical examples are the Loitsyanskii invariant in homogeneous isotropic turbulence (Loitsyanskii 1939) and the Corrsin invariant for a homogeneous isotropic contaminant field (Corrsin 1951). Generalizations of these invariants to anisotropic fields are given by Monin & Yaglom (1975, pp. 149–152), and other invariants are derived by Lumley (1966).

During recent investigations, described in Chatwin & Sullivan (1979*a*, *b*), the authors discovered that there are certain invariant properties that follow directly from (1.4). The purpose of this paper is to derive these properties and to discuss, with experimental support, some of their applications and implications.

2. The invariant properties

Denote ensemble means by overbars and write

$$\Gamma(\mathbf{x},t) = C(\mathbf{x},t)+c(\mathbf{x},t), \quad C = \overline{\Gamma}, \quad \bar{c} = 0, \tag{2.1}$$

so that $C(\mathbf{x},t)$ is the ensemble mean concentration and $c(\mathbf{x},t)$ is the fluctuation. In view of the essential inhomogeneity and non-stationarity of the statistical properties of $\Gamma(\mathbf{x},t)$ and the (unjustified) unfamiliarity of relative diffusion, it is useful, for clarity, to discuss very briefly the evaluation of ensemble means. The basic point is that the definition of $\mathbf{x}$ requires that ensemble means, derived from the set of realizations of $\Gamma(\mathbf{x},t)$, be evaluated only after the centres of mass of all realizations of the cloud have been made coincident for each t. Thus suppose that in an experimental investigation N realizations of the dispersion are performed. Let

$$\Gamma_0^{(n)}(\mathbf{X},t)\;(n = 1,2,\ldots,N)$$

be the distribution of concentration in the nth realization so that (1.1) is satisfied for each n. Then define $\mathbf{X}_c^{(n)}(t)$ and $\Gamma^{(n)}(\mathbf{x},t)$ by the obvious extensions of (1.2) and (1.3), *viz.*

$$Q\mathbf{X}_c^{(n)}(t) = \int \mathbf{X}\Gamma_0^{(n)}(\mathbf{X},t)\,dV(\mathbf{X}), \quad \Gamma^{(n)}(\mathbf{x},t) = \Gamma_0^{(n)}(\mathbf{x}+\mathbf{X}_c^{(n)},t). \tag{2.2}$$

Then ensemble means of functions of $\Gamma(\mathbf{x},t)$ are determined in the normal way, typified by the following result for the ensemble mean concentration $C(\mathbf{x},t)$:

$$C(\mathbf{x},t) = \lim_{N\to\infty}\left\{N^{-1}\sum_{n=1}^{N}\Gamma^{(n)}(\mathbf{x},t)\right\}. \tag{2.3}$$

That this discussion is not entirely trivial is clear when it is realized for example that $C(\mathbf{x},t)$ is not the same as $\overline{\Gamma}_0(\mathbf{x}+\overline{\mathbf{X}}_c,t)$.†

From (1.4) and (2.2) it follows that, for each n,

$$\int \Gamma^{(n)}(\mathbf{x},t)\,dV(\mathbf{x}) = Q, \quad \int \mathbf{x}\Gamma^{(n)}(\mathbf{x},t)\,dV(\mathbf{x}) = 0,$$

and so, from (2.1) and (2.3), that

$$\int C(\mathbf{x},t)\,dV(\mathbf{x}) = Q, \quad \int c(\mathbf{x},t)\,dV(\mathbf{x}) = 0, \tag{2.4}$$

and that

$$\int \mathbf{x}C(\mathbf{x},t)\,dV(\mathbf{x}) = 0, \quad \int \mathbf{x}c(\mathbf{x},t)\,dV(\mathbf{x}) = 0. \tag{2.5}$$

The statistical property of special interest here is $r(\mathbf{y},t)$ defined by

$$r(\mathbf{y},t) = Q^{-2}\int \overline{c(\mathbf{x},t)\,c(\mathbf{x}+\mathbf{y},t)}\,dV(\mathbf{x}). \tag{2.6}$$

As pointed out in Chatwin & Sullivan (1978, 1979*b*) r is a correlation function (in the sense normal in turbulence theory) whose Fourier transform is the energy spectrum of $\int \overline{c^2}(\mathbf{x},t)\,dV(\mathbf{x})$. In the present paper it is more important that $r(\mathbf{y},t)$ is one of two main contributions to the distance-neighbour function.

The invariants referred to in the title of this paper are invariants of $r(\mathbf{y},t)$. It is immediate from (2.4) and (2.6) that

$$r(\mathbf{y},t) \equiv r(-\mathbf{y},t) \quad \text{for all} \quad \mathbf{y}; \tag{2.7}$$

and that

$$\int r(\mathbf{y},t)\,dV(\mathbf{y}) = 0. \tag{2.8}$$

Also define $m^{(n)}_{ij\ldots k}(t)$ by

$$m^{(n)}_{ij\ldots k}(t) = \int y_i y_j \ldots y_k r(\mathbf{y},t)\,dV(\mathbf{y}), \tag{2.9}$$

where n is the number of tensor suffices. It follows from (2.7) that

$$m^{(2n+1)}_{ij\ldots k}(t) = 0 \quad \text{for all} \quad n, \tag{2.10}$$

and in particular that

$$m^{(1)}_i(t) = \int y_i r(\mathbf{y},t)\,dV(\mathbf{y}) = 0. \tag{2.11}$$

† Consider, for example, the simple (though highly artificial) case when the motion of the centre of mass is random, but there is no dispersion about the centre of mass (a 'billiard-ball' cloud). Then $C(\mathbf{x}, t)$ is constant and equal to the initial concentration, but $\Gamma_0(\mathbf{x}+\mathbf{X}_c, t)$ cannot be (even if $\mathbf{X}_c = 0$) whenever $\mathbf{X}_c^2 \neq 0$.

It is interesting to note here that (2.7) to (2.11) have been derived without use of (2.5), the condition distinguishing relative from absolute diffusion. Therefore (2.7) to (2.11) hold in absolute diffusion, when $r(\mathbf{y},t)$ is defined by an equation like (2.6) with $\Gamma_0(\mathbf{X},t)-\overline{\Gamma}_0(\mathbf{X},t)$ replacing $c(\mathbf{x},t)$, etc. Another interesting point is to compare (2.7) and (2.8) with the generalizations of Corrsin's invariant referred to above. That work considers a homogeneous contaminant field which means that the statistical properties of $\Gamma_0(\mathbf{X},t)$ are independent of $\mathbf{X}$. In those circumstances $r(\mathbf{y},t)$ cannot be defined by an equation like (2.6) since the integral over all space cannot exist. The natural correlation to use in that case is $r_*(\mathbf{y},t)$ defined by

$$r_*(\mathbf{y},t) = \overline{c_0(\mathbf{X},t)\,c_0(\mathbf{X}+\mathbf{y},t)}, \tag{2.12}$$

where

$$c_0(\mathbf{X},t) = \Gamma_0(\mathbf{X},t)-\overline{\Gamma}_0(\mathbf{X},t). \tag{2.13}$$

It follows from homogeneity that

$$r_*(\mathbf{y},t) \equiv r_*(-\mathbf{y},t) \quad \text{for all} \quad \mathbf{y}, \tag{2.14}$$

which is to be compared with (2.7). It can also be shown from the diffusion equation governing $\Gamma_0(\mathbf{X},t)$ that

$$\int r_*(\mathbf{y},t)\,dV(\mathbf{y}) = \text{constant}, \tag{2.15}$$

(Corrsin 1951; Monin & Yaglom 1975, p. 150). While $r(\mathbf{y},t)$ and $r_*(\mathbf{y},t)$ seem to be the simplest, and most natural, correlation functions† for the respective situations which they describe, and while they both have a constant integral, there is no evidence that the constant in (2.15) is generally zero, unlike that in (2.8). It is also interesting to note that, while Corrsin's original derivation of (2.15) was from the diffusion equation, his result can be obtained, like (2.8), using only conservation of mass – see Monin & Yaglom (1975, p. 152).

The final invariant property to be presented here is that

$$m_{ij}^{(2)}(t) = \int y_i y_j r(\mathbf{y},t)\,dV(\mathbf{y}) = 0. \tag{2.16}$$

Unlike (2.7) to (2.11) this holds only in relative diffusion, so that (2.5) is needed. The proof of (2.16) is easy. By the definition of $r(\mathbf{y},t)$ in (2.6),

$$\begin{aligned} m_{ij}^{(2)}(t) &= Q^{-2}\int\int y_i y_j \overline{c(\mathbf{x},t)\,c(\mathbf{x}+\mathbf{y},t)}\,dV(\mathbf{x})\,dV(\mathbf{y}),\\ &= Q^{-2}\int\int (z_i-x_i)(z_j-x_j)\,\overline{c(\mathbf{x},t)\,c(\mathbf{z},t)}\,dV(\mathbf{x})\,dV(\mathbf{z}),\\ &= 2Q^{-2}\int\int x_i x_j \overline{c(\mathbf{x},t)\,c(\mathbf{z},t)}\,dV(\mathbf{x})\,dV(\mathbf{z})\\ &\quad -2Q^{-2}\overline{\left\{\int x_i c(\mathbf{x},t)\,dV(\mathbf{x})\right\}\left\{\int x_j c(\mathbf{x},t)\,dV(\mathbf{x})\right\}}. \end{aligned}$$

† Note that although r_* can be defined by (2.12) for the finite cloud considered in this paper, it depends on $\mathbf{X}$, as well as $\mathbf{y}$ and t. This is also true when an r_* is defined in terms of $c(\mathbf{x},t)$. On the other hand, as noted earlier, r does not exist for a homogeneous contaminant field.

The integral over $\mathbf{z}$ in the first term vanishes by (2.4), and each of the two integrals in the second term vanishes by (2.5). It is perhaps worth reiteration that (2.16) requires no assumption about the contaminant or the turbulent velocity, other than that the total mass be conserved and that the statistical properties be defined in the framework of relative diffusion.

It is evident that many other statistical properties (defined, for example, in terms of the values of $c(\mathbf{x}, t)$ at more than two points) will have invariants derived by use of (2.4) and (2.5). This remark will not be taken further here.

3. Some consequences of the invariants

The analogous results for a plume and experimental support

Experiments on clouds are rare because of their obvious difficulty. However analogous invariants hold for steady plumes on which observations are much more commonly made. To be specific, consider the experimental situation used by Sullivan (1965), and described in Sullivan (1971), Chatwin & Sullivan (1979*b*). Dye was released at a steady rate from a source in the thin surface layer of Lake Huron, and was advected downstream at the surface water velocity (more precisely by the combined effects of the water velocities in the thin surface layer). Essentially therefore the dye dispersed in two dimensions. Let x denote distance from the source along the instantaneous centre-line of the dye plume, and let y denote distance in the lake surface normal to this centre line. Define $\Gamma(y, x, t)$ and $c(y, x, t)$ in the ways obviously analogous to those described above for a cloud (Chatwin & Sullivan 1979*b*), and $r(y, x)$ by the analogue to (2.6), namely

$$r(y,x) = Q^{-2}\int_{-\infty}^{\infty} \overline{c(z,x,t)\,c(z+y,x,t)}\,dz, \tag{3.1}$$

where Q is a measure of the rate of dye emission by the source, and r does not depend on t because this rate is constant.

Using approximations derived in § 3 of Chatwin & Sullivan (1979*b*) – in particular equation (3.14) of that paper – it is easy to show that the analogues of (2.7) to (2.11), and (2.16) hold approximately. Thus

$$r(y,x) \equiv r(-y,x) \quad \text{for all} \quad y; \tag{3.2}$$

$$m^{(0)}(x) = \int_{-\infty}^{\infty} r(y,x)\,dy \approx 0; \tag{3.3}$$

$$m^{(2n+1)}(x) = \int_{-\infty}^{\infty} y^{2n+1} r(y,x)\,dy = 0; \tag{3.4}$$

$$m^{(2)}(x) = \int_{-\infty}^{\infty} y^{2} r(y,x)dy \approx 0. \tag{3.5}$$

Of these results, only (3.5) requires the use of a relative diffusion framework (as with the analogous result (2.16) for a cloud).

Table 1 gives values of $m^{(0)}$ and $m^{(2)}$ for the plumes in Sullivan's experiments. Within experimental accuracy both are zero.

The representation of $r(y,x)$ and $r(\mathbf{y},t)$

It is conventional in turbulence theory to represent random functions and correlation functions like $r_*(\mathbf{y},t)$ in terms of Fourier transforms. This is appropriate when the random function is stationary with respect to the transformed variable (Monin & Yaglom 1975, pp. 1–22), but not when it is inherently non-stationary or inhomogeneous. In the case of the steady plume considered above, both $\Gamma(y,x,t)$ and $c(y,x,t)$ are inhomogeneous in y because of the requirement that there be constant finite mass flux along the plume. Observations suggest (Sullivan 1971; Chatwin & Sullivan 1979*b*) that the statistical properties decay to zero as $y \to \pm\infty$ like $\exp(-\frac{1}{2}\alpha y^2)$ for some α. Such rapid decay ought, it seems, to be reflected in a choice of functions other than $\exp(iky)$ for the representation of $\Gamma(y,x,t)$ and $c(y,x,t)$.

A set of complete functions on $(-\infty,\infty)$ is $H_n(y)\exp(-\frac{1}{2}y^2)$ $(n = 0, 1, 2, \ldots)$, where $H_n(y)$ is the nth Hermite polynomial defined by

$$H_n(y)\exp(-\tfrac{1}{2}y^2) = (-1)^n (d/dy)^n \exp(-\tfrac{1}{2}y^2). \tag{3.6}$$

The use of these functions in turbulence theory has been discussed by Kampé de Fériet (1966) and Lumley (1970). Whether they can profitably be used to represent random functions like $\Gamma(y,x,t)$ or $c(y,x,t)$ will not be discussed here (although they were employed by Chatwin (1970) in a discussion of turbulent diffusion of a cloud in a pipe). But consider $r(y,x)$ defined by (3.1). For any $l(x)$, it can be represented formally in the following way:

$$r(y,x) = \sum_{n=0}^{\infty} A_n(x)\,H_n(Y)\exp(-\tfrac{1}{2}Y^2), \quad Y = \frac{y}{l(x)}, \tag{3.7}$$

where the $A_n(x)$ are obtained by

$$A_n(x) = \frac{1}{2\pi n!}\int_{-\infty}^{\infty} r(y,x)\,H_n(Y)\,dY. \tag{3.8}$$

(The question of the conditions on $r(y,x)$ which guarantee convergence of the series in (3.7) is discussed by Kendall & Stuart (1969, pp. 161–163), but it may be sufficient if the series is simply asymptotic, or convergent in a generalized sense.) Because of the form of the Hermite polynomials, the invariant relations (3.2) to (3.5) ensure that $A_{2n+1}(x) = 0$ $(n = 0, 1, \ldots)$ and that $A_0(x) = A_2(x) = 0$. Thus (3.7) reduces to

$$r(y,x) = \{A_4(x)\,H_4(Y) + A_6(x)\,H_6(Y) + \ldots\}\exp(-\tfrac{1}{2}Y^2).\dagger \tag{3.9}$$

That this representation is useful practically is shown in figure 1 where the solid line shows $r(y,x)$ measured in the experiments by Sullivan (1965) and presented in Chatwin & Sullivan (1979*b*),‡ and the dashed line shows the first term of (3.9); the curves were normalized to coincide at $y = Y = 0$ and $l(x)$ was chosen to be $(\frac{10}{9})\,L(x)$, where $L(x)$ was a mean plume width defined and measured in Chatwin & Sullivan (1979*b*). Considering the experimental errors involved, the agreement is excellent. It should be noted that the widths of the plumes in the experiments were such that they lay within Kolmogoroff's inertial subrange; the factor $(\frac{10}{9})$ used to fit the theory with

† Since r is a true correlation function, its Fourier transform must be non-negative for all Y; this is satisfied for (3.9) provided $(-1)^n A_{2n}(x) \geqslant 0$.

‡ In the notation of Chatwin & Sullivan (1979*b*) the curve shown here is $Q_1(Y)$.

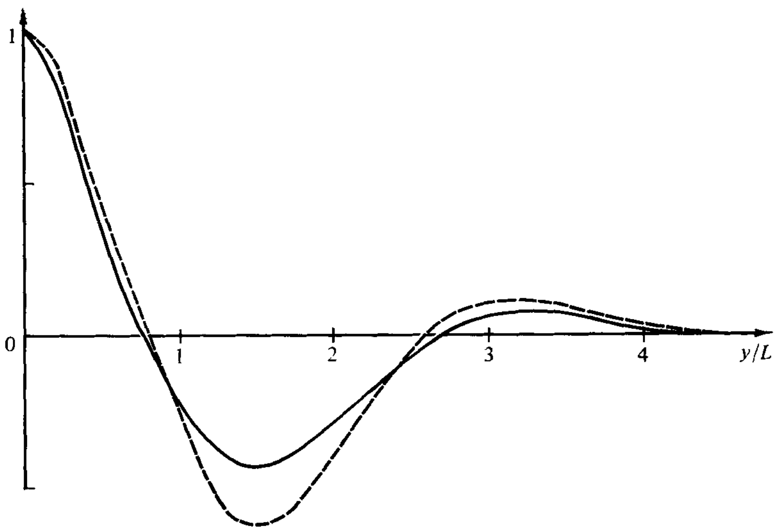

FIGURE 1. The solid line is $r(y, x)$ defined in (3.1) and measured by Chatwin & Sullivan (1979*b*) The dashed line is the first term of (3.9).

the data should then be independent of Reynolds number (provided a precise criterion is used for defining $l(x)$ – unlike the situation in this paper!). In general the appropriate factor could depend on the injection conditions, as well as Reynolds number, if this were too small for Kolmogoroff's theory to be applicable.

The experimental curve in figure 1 was determined from twelve sets of data in the way described in detail by Sullivan (1971) and Chatwin & Sullivan (1979*b*). Table 1 shows the values of certain normalized moments $\mu^{(n)}(x)$ of $r(y,x)$ for each set of data, where

$$2\mu^{(n)}(x) = m^{(n)}(x)\frac{\int_{-\infty}^{\infty} C(y,x)\,dy}{\int_{-\infty}^{\infty} y^n C(y,x)\,dy}. \tag{3.10}$$

If $r(y,x)$ is described by the first term of (3.9) and if, as Sullivan's observations suggest, $C(y,x)$ is a Gaussian function of y, then it is easily shown that

$$\mu^{(n)}(x) = \frac{Lr(0,x)\,(2\pi)^{\frac{1}{2}}}{6}\left(\frac{l}{L}\right)^{n+1} n(n-2). \tag{3.11}$$

The dependence of $\mu^{(n)}(x)$ on $L(x)$ and $r(0,x)$ is discussed later. To eliminate this, table 1 shows also the measured values of $\mu^{(6)}/\mu^{(4)}$, $\mu^{(8)}/\mu^{(6)}$ and $\mu^{(8)}/\mu^{(4)}$. Within experimental error, and given the limited number of realizations, the median values of these ratios are not inconsistent with the values predicted by (3.11). This is further support that, in practice, the first term of (3.9) may well describe $r(y,x)$.

A description of the relative dispersion of a plume was given in Chatwin & Sullivan (1979*b*) based on the fact that significant values of $c(y,x,t)$ are generated by the transfer of contaminant from the central region of the plume outwards by those eddies primarily responsible for the plume spread. On the basis of this idea of a dominant eddy size – of length scale of order $L(x)$ – it was argued that $r(y,x)$ should have

Station	Number of crossings	$m^{(0)}/m_*^{(0)}$	$m^{(2)}/m_*^{(2)}$	$\mu^{(4)}$	$\mu^{(6)}$	$\mu^{(8)}$	$\mu^{(6)}/\mu^{(4)}$	$\mu^{(8)}/\mu^{(6)}$	$\mu^{(8)}/\mu^{(4)}$	L
1–1	25	0·016	0·003	0·121	0·174	1·19	1·4	6·9	10·0	10·9
1–2	8	0·011	0·036	0·200	0·740	1·96	3·7	2·7	10·0	12·7
2–1	25	0·450	0·395	0·100	0·990	3·55	9·9	3·6	35·6	1·6
2–2	20	0·018	0·005	0·128	0·470	1·45	3·7	3·1	11·2	12·2
3–1U	26	0·050	0·036	1·26	6·70	24·0	5·3	3·6	19·2	3·5
3–1L	27	0·067	0·264	0·173	1·10	3·73	6·3	3·4	21·6	2·1
3–2U	16	0·030	0·026	0·218	0·995	3·14	4·5	3·1	14·4	5·2
3–2L	16	0·042	0·049	0·175	0·795	2·47	4·6	3·1	14·4	3·9
4–1U	12	0·020	0·036	0·032	0·040	0·217	1·3	5·5	6·8	7·8
4–1L	9	0·048	0·059	0·283	1·14	2·82	4·1	2·5	10·0	3·9
4–2	10	0·010	0·002	0·057	0·165	2·42	2·9	1·5	4·4	14·4
5–1	15	0·017	0·012	0·165	0·875	3·20	5·3	3·7	19·2	8·3
				Median value with $l/L = \frac{10}{9}$			4·1	3·1	11·2	
				Value from (3.11)			3·7	2·5	9·2	

TABLE 1. The values of the moments of $r(y, x)$. For specification of the stations, the first number refers to the plume, the second to the downstream location and the letters U or L to upper or lower reading levels (see Chatwin & Sullivan 1979*b*). The moments $m^{(n)}$, $\mu^{(n)}$ are defined in (3.4), (3.10) and $m_*^{(n)} = \int_{-\infty}^{\infty} y^n\,|r(y,x)|\,dy$. One unit of L is 3·36 m.

zeros at $y = O(L)$ and $y = O(3L)$ with stationary values at $y = O(2L)$ and $y = O(4L)$. Now $H_4(Y)\exp(-\frac{1}{2}Y^2)$ has its positive zeros at 0·74 and 2·33, and its positive stationary values at 1·36 and 2·86, and these are approximately in the ratio 1 : 3 : 2 : 4 predicted by the simple description. This suggests both that the description is reasonable and that $r(y, x)$ can be approximated well by the first term of (3.9), as figure 1 and table 1 confirm.

The one-dimensional representation (3.9) cannot be used for the cloud correlation function $r(\mathbf{y}, t)$. But generalizations of Hermite polynomials to more than one dimension exist (Kampé de Fériet 1966). Formally $r(\mathbf{y}, t)$ can be represented in the following way for an arbitrary positive definite tensor $l_{pq}(t)$:

$$r(\mathbf{y},t) = \sum_{n=0}^{\infty} A^{(n)}_{ij\ldots k}(t)\, H^{(n)}_{ij\ldots k}(\mathbf{y},t)\exp(-\tfrac{1}{2}l_{pq}y_p y_q), \tag{3.12}$$

where the summation convention is used with $i, j, \ldots, k$, as earlier n is the number of tensor suffices, and $H^{(n)}_{ij\ldots k}(\mathbf{y}, t)$ is defined by

$$H^{(n)}_{ij\ldots k}(\mathbf{y},t)\exp(-\tfrac{1}{2}l_{pq}y_p y_q) = (-1)^n \frac{\partial^n}{\partial y_i\,\partial y_j \ldots \partial y_k}\exp(-\tfrac{1}{2}l_{pq}y_p y_q). \tag{3.13}$$

The $A^{(n)}_{ij\ldots k}(t)$ can be obtained from integrals generalizing (3.8) which will not be written down here. (But note that these integrals now involve polynomials $G^{(n)}_{ij\ldots k}(\mathbf{y}, t)$ defined by an equation like (3.13) but replacing l_{pq} by its inverse. Details and further references are given in Kampé de Fériet (1966).) It is sufficient to note here that the invariant relations (2.7) to (2.11), and (2.16), once more require all the odd terms in (3.12) to vanish identically, and also require $A^{(0)}(t) = A^{(2)}_{ij}(t) = 0$. Thus, as in (3.9), the expansion (3.12) involves only even terms and begins with $n = 4$. No observations exist to test this result, but it should be practically useful.

Relations between moments of $r(\mathbf{y}, t)$, Γ *and the distance-neighbour function*

The moments $m^{(n)}_{ij\ldots k}(t)$ of $r(\mathbf{y}, t)$ defined in (2.9) can be related to the moments of $c(\mathbf{x}, t)$. It is easily shown, for example, that $m^{(4)}_{ijkl}(t)$, the first moment of $r(\mathbf{y}, t)$ that is not identically zero, satisfies

$$m^{(4)}_{ijkl}(t) = 2\{\overline{b^{(2)}_{ij}\, b^{(2)}_{kl}} + \overline{b^{(2)}_{ik}\, b^{(2)}_{jl}} + \overline{b^{(2)}_{il}\, b^{(2)}_{jk}}\}, \tag{3.14}$$

where

$$b^{(2)}_{ij}(t) = Q^{-1}\int x_i x_j c(\mathbf{x}, t)\, dV(x). \tag{3.15}$$

In particular

$$m^{(4)}_{iijj}(t) = \int |\mathbf{y}|^4 r(\mathbf{y}, t)\, dV(\mathbf{y}) = 2\{\overline{b^{(2)}_{ii} b^{(2)}_{jj}}\} + 4\{\overline{b^{(2)}_{ij} b^{(2)}_{ij}}\},$$

and when the turbulence is isotropic and the initial distribution of contaminant is spherically symmetric,

$$m^{(4)}_{ijkl}(t) = \tfrac{1}{15} m^{(4)}_{ppqq}(t)\,\{\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\}.$$

Of more direct physical and practical importance are the relationships between the distance-neighbour function $p(\mathbf{y}, t)$ and the statistical properties of Γ, where $p(\mathbf{y}, t)$ is defined by

$$p(\mathbf{y}, t) = Q^{-2}\int \overline{\Gamma(\mathbf{x}, t)\,\Gamma(\mathbf{x}+\mathbf{y}, t)}\, dV(\mathbf{x}). \tag{3.16}$$

This definition is essentially that of Batchelor (1952). Using (2.1) it follows that

$$p(\mathbf{y}, t) = Q^{-2}\int C(\mathbf{x}, t)\, C(\mathbf{x}+\mathbf{y}, t)\, dV(\mathbf{x}) + r(\mathbf{y}, t). \tag{3.17}$$

(Note that while it is clear that the same value of $p(\mathbf{y}, t)$ is obtained if $\Gamma(\mathbf{x}, t)$ and $\Gamma(\mathbf{x}+\mathbf{y}, t)$ in (3.16) are replaced by $\Gamma_0(\mathbf{X}, t)$ and $\Gamma_0(\mathbf{X}+\mathbf{y}, t)$, each of the two terms on the right-hand side of (3.17) is then different although their sum is of course unchanged.)

Define $M^{(n)}_{ij\ldots k}(t)$ and $B^{(n)}_{ij\ldots k}(t)$ by

$$M^{(n)}_{ij\ldots k}(t) = \int y_i y_j \ldots y_k p(\mathbf{y}, t)\, dV(\mathbf{y}), \quad B^{(n)}_{ij\ldots k}(t) = Q^{-1}\int x_i x_j \ldots x_k C(\mathbf{x}, t)\, dV(\mathbf{x}). \tag{3.18}$$

It is easy to show from (3.16) that $p(\mathbf{y}, t)$ is even in $\mathbf{y}$, so that

$$M^{(2n+1)}_{ij\ldots k}(t) = 0 \quad \text{for all} \quad n. \tag{3.19}$$

Using (2.8) and (2.16) it follows from (2.4), (2.5) and (3.17) that

$$M^{(0)}(t) = 1, \quad M^{(2)}_{ij}(t) = 2B^{(2)}_{ij}(t), \tag{3.20}$$

(Batchelor 1952). More generally it is fairly easy to show that for each $n \geqslant 2$,

$$\{M^{(2n)}_{ij\ldots k} - m^{(2n)}_{ij\ldots k}\}$$

is a function only of the values of $B^{(r)}_{ij\ldots k}$ for $r \leqslant 2n$. For $n = 2$ the relationship is

$$M^{(4)}_{ijkl} - m^{(4)}_{ijkl} = 2\{B^{(4)}_{ijkl} + B^{(2)}_{ij} B^{(2)}_{kl} + B^{(2)}_{ik} B^{(2)}_{jl} + B^{(2)}_{il} B^{(2)}_{jk}\}. \tag{3.21}$$

Similar results to these hold for steady plumes. Thus for the geometry considered earlier define $p(y,x)$ by

$$p(y,x) = Q^{-2}\int \overline{\Gamma(z,x,t)\,\Gamma(z+y,x,t)}\,dz = Q^{-2}\int C(z,x)\,C(z+y,x)\,dz + r(y,x), \quad (3.22)$$

where it is convenient to choose Q to be equal to the uniform value of $\int_{-\infty}^{\infty} C(z,x)\,dz$. Since $p(y,x)$ is even in y all its odd moments vanish identically. Others results can be derived in an analogous manner to those above. With an obvious modification to the notation, some of these are:

$$M^{(0)}(x) = \int_{-\infty}^{\infty} p(y,x)\,dy = 1; \quad (3.23)$$

$$M^{(2)}(x) = \int_{-\infty}^{\infty} y^2 p(y,x)\,dy = 2Q^{-1}\int_{-\infty}^{\infty} z^2 C(z,x)\,dz = 2B^{(2)}(x); \quad (3.24)$$

$$M^{(4)}(x) - m^{(4)}(x) = 2B^{(4)}(x) + 6\{B^{(2)}(x)\}^2; \quad (3.25)$$

$$M^{(6)}(x) - m^{(6)}(x) = 2B^{(6)}(x) + 30B^{(4)}(x)\,B^{(2)}(x) - 20\{B^{(3)}(x)\}^2. \quad (3.26)$$

The terms on the right-hand sides of (3.23) to (3.26) are those given by Richardson (1926). However he did not define his distance-neighbour function as an ensemble average, so the terms $m^{(2n)}(x)$ coming from the fluctuations were not present on the left-hand sides of these equations.

It is clear from these relations however that, if the moments $m^{(n)}$ of $r(\mathbf{y},t)$ or $r(y,x)$ all vanish, the moments of the distance-neighbour function can be determined from those of C, and, if C is even, its moments can be determined from those of the distance–neighbour function. Normally a function can be determined uniquely if all of its moments are known (Monin & Yaglom 1971, pp. 225–226), so that it is of interest to ask whether the moments of $r(\mathbf{y},t)$ or $r(y,x)$ ever vanish identically, or, more realistically, are small compared with those of the distance–neighbour function.

Figure 1 shows that within experimental error $r(y,x)$ can be well approximated by the first term in (3.9). Since $H_4(0) = 3$ this can be written

$$r(y,x) \approx \tfrac{1}{3} r(0,x)\,H_4(Y)\exp(-\tfrac{1}{2}Y^2), \quad (3.27)$$

where, by (3.1),

$$r(0,x) = Q^{-2}\int_{-\infty}^{\infty} \overline{c^2(z,x,t)}\,dz. \quad (3.28)$$

The right-hand side of (3.28) was measured in Chatwin & Sullivan (1979*b*) and found to be proportional to $\{L(x)\}^{-1.5}$, where $L(x)$ satisfies

$$QL^2(x) = \int_{-\infty}^{\infty} z^2 C(z,x)\,dz. \quad (3.29)$$

This measured decay of $r(0,x)$ with $L(x)$ is consistent within experimental error with a least squares fit to the measured values of $\mu^{(n)}(x)$, defined in (3.11) and given in table 1. On the other hand, (3.23) requires $p(y,x)$ to be proportional to $\{L(x)\}^{-1}$ so that

$$r(y,x)/p(y,x) \propto \{L(x)\}^{-0.5}, \quad (3.30)$$

in these experiments. Since the Reynolds number in the experiments was large, it follows that

$$L(x) \approx A(\epsilon x^3/U^3)^{\frac{1}{2}}, \tag{3.31}$$

where U is the mean downstream speed, ϵ is the rate of dissipation of mechanical energy per unit mass and A is a constant. The conclusion is that the moments $m^{(n)}(x)$ of $r(y,x)$ become increasingly negligible as $x \to \infty$ compared with the moments $M^{(n)}(x)$ of the distance–neighbour function $p(y,x)$. It then becomes possible for the moments of $C(y,x)$ to determine uniquely all the moments of $p(y,x)$, and *vice-versa*, if $C(y,x)$ is even in y.

The results of the experiments used to derive (3.30) showed (Sullivan 1971) that in them $C(y,x)$ and $p(y,x)$ both approached Gaussian functions of y, the approach in the case of C being the faster (Sullivan 1975). While it does not seem possible to prove rigorously, using the ideas and techniques described in Lumley (1972), that this behaviour occurs, it is not inconsistent with other observations (Monin & Yaglom 1975, pp. 577–578). Sullivan (1976) suggested that a necessary condition for $p(y,x)$ to be Gaussian was that x be greater than about $300U(\nu/\epsilon)^{\frac{1}{2}}$.

It seems likely that similar conclusions can be made for a cloud although, as explained earlier, no experimental work is available.

The decay with x of the right-hand side of (3.28), and the decay with t of the equivalent expression for a cloud depend on κ, the molecular diffusivity, in the sense that, if $\kappa = 0$, no decay occurs. Note that without this decay $p(0,x)$ and $p(0,t)$ would be constant. It follows that theoretical discussions of clouds or plumes of marked fluid *particles* (i.e. turbulent diffusion with $\kappa, = 0$) could not determine C in terms of p (or *vice-versa*), nor explain the approach to Gaussianity of either. The point is that r is definitely not Gaussian as the analysis in this paper shows, and does not decay if $\kappa = 0$. The mechanism of the decay is discussed in Chatwin & Sullivan (1979*a*, *b*).

Paul J. Sullivan acknowledges the financial support of the National Research Council of Canada during the period when this work was carried out.

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