J. Fluid Mech. (1972), *vol.* 51, *part* 1, *pp.* 63–67
Printed in Great Britain

The cumulants of the distribution of concentration of a solute dispersing in solvent flowing through a tube

By P. C. CHATWIN
Department of Applied Mathematics,
The University of Liverpool

(Received 20 May 1971)

The mean distribution of concentration of a solute over the cross-section of a tube through which a solvent is flowing is given approximately by the solution of an equation of the heat-conduction type (Taylor 1953, 1954). One solution of this equation is a Gaussian curve but observed distributions are normally not Gaussian. On the basis of an equation derived by Chatwin (1970) it is shown here that the deviation of an observed distribution from the Gaussian curve with the same mean and variance is not determined by the heat-conduction equation, although the practical importance of this remark may be small if the initial distribution of solute is greatly elongated along the tube axis.

1. Introduction

When a solute is injected into a tube through which solvent is flowing it spreads out along the axis, Ox say, under the combined effects of advection with the solvent, and molecular and/or turbulent diffusion. If this diffusion can be described by the gradient law, if the tube has constant cross-section and if the solute can be regarded as a passive marker then, for large values of the time t, the mean concentration of solute over the cross-section C_m is approximately given by the solution of a simple equation of the heat-conduction type (Taylor 1953, 1954). This equation can be written

$$\frac{\partial C_m}{\partial t} + U\frac{\partial C_m}{\partial x} = K_2\frac{\partial^2 C_m}{\partial x^2}, \tag{1.1}$$

where U is the discharge velocity and K_2 is a constant which depends on the velocity profile and the mechanics of the diffusion process. For a cloud of solute of finite extent in which (1.1) holds, it is easy to show that the centre of mass x_g and the variance σ^2 of the cloud have rates of change U and $2K_2$ respectively and it is possible, and convenient, to choose the origins of x and t (the latter may be virtual) so that

$$x_g = Ut, \quad \sigma^2 = 2K_2 t. \tag{1.2}$$

It is also convenient to choose units of concentration so that

$$\int_{-\infty}^{\infty} C_m dx = 1. \tag{1.3}$$

Now one solution of (1.1) is the Gaussian curve

$$C_m(x,t) = (1/\sigma(2\pi)^{\frac{1}{2}})\exp\{-(x-x_g)^2/2\sigma^2\}, \tag{1.4}$$

which has the property that all its cumulants of order greater than two vanish. The general solution of (1.1) does not have this property, nor do observed distributions of C_m(except as $t \to \infty$). This remark suggests the following question which was put to me by Professor H. B. Fischer. Can the evolution of observed, but non-Gaussian, distributions of C_m be described by more general solutions of (1.1) than (1.4) with greater accuracy than that given by (1.4)? This note shows that the answer to this question is no, except possibly for special initial distributions. The reason for this is that (1.1) is approximate and that the degree of approximation is such that the cumulants of order greater than two are not in general given, even approximately, by its general solution.

2. The values of the absolute cumulants of C_m for large times

It is shown in Chatwin (1970) that, formally, C_m satisfies

$$\frac{\partial C_m}{\partial t} + U\frac{\partial C_m}{\partial x} = K_2\frac{\partial^2 C_m}{\partial x^2} + K_3\frac{\partial^3 C_m}{\partial x^3} + K_4\frac{\partial^4 C_m}{\partial x^4} + \ldots, \tag{2.1}$$

as $t \to \infty$, where K_3, K_4, ... are constants depending, like K_2, on the particular velocity profiles and diffusion processes in the flow. Equation (1.1) is obtained from (2.1) by neglecting all terms except the first one on the right-hand side. It is easy to show, using the moment method of Aris (1956), that the terms in (2.1) involving $K_3, K_4, \ldots$, do not affect x_g and σ^2, so that the results in (1.2) are still valid. The differences between (2.1) and (1.1) only appear when other shape parameters like the cumulants of order greater than two are considered.

It is neatest to work with the following variables:

$$X = (x-x_g)/\sigma, \quad p(X,\sigma) = \sigma C_m. \tag{2.2}$$

Thus X is the 'standard measure' and, from (1.3),

$$\int_{-\infty}^{\infty} p(X,\sigma)dX = 1.$$

If $\phi(k,\sigma)$ is defined by

$$\phi(k,\sigma) = \int_{-\infty}^{\infty} p(X,\sigma)e^{ikX}\,dX,$$

then, by definition (Kendall & Stuart 1958, pp. 67–68),

$$\Phi(k,\sigma) = \log_e \phi(k,\sigma) = -\tfrac{1}{2}k^2 + \sum_{n=3}^{\infty}\frac{(ik)^n}{n!}\lambda_n(\sigma), \tag{2.3}$$

where λ_n is the nth absolute cumulant of C_m. In writing (2.3) the facts that $\lambda_1 = 0$ and $\lambda_2 = 1$, which follow from (1.2), have been used.

Equation (2.1) can be transformed into an equation for p in terms of X and σ. When the Fourier transform of this equation with respect to X is taken the resulting equation can be written (after some algebra and integrations by parts) as

$$\sigma\frac{\partial\Phi}{\partial\sigma}+k\frac{\partial\Phi}{\partial k} = (ik)^2-\frac{(ik)^3}{\sigma}\frac{K_3}{K_2}+\frac{(ik)^4}{\sigma^2}\frac{K_4}{K_2}\cdots$$

This equation can be integrated by using a new pair of dependent variables, ik/σ and ik for example. It then follows that its general solution is

$$\Phi = f\left(\frac{ik}{\sigma}\right)+\frac{1}{2}\left[(ik)^2-\frac{(ik)^3}{\sigma}\frac{K_3}{K_2}+\frac{(ik)^4}{\sigma^2}\frac{K_4}{K_2}\cdots\right], \tag{2.4}$$

where f is any function whose Taylor series begins, for consistency with (2.3), with the term in $(ik/\sigma)^3$.

On comparing coefficients of $(ik)^n$ in (2.3) and (2.4) it follows that as $t\rightarrow\infty$ and for $n \geqslant 3$,

$$\lambda_n = \tfrac{1}{2}(-1)^n n!\,(K_n/K_2)\sigma^{2-n}+O(\sigma^{-n}). \tag{2.5}$$

In particular λ_3 and λ_4, which are commonly used as measures of skewness and kurtosis respectively, satisfy

$$\left.\begin{aligned}\lambda_3 &= -3(K_3/K_2)\sigma^{-1}+O(\sigma^{-3}) = -\tfrac{3}{2}\sqrt{2}K_3K_2^{-\frac{3}{2}}t^{-\frac{1}{2}}+O(t^{-\frac{3}{2}}),\\ \lambda_4 &= 12(K_4/K_2)\sigma^{-2}+O(\sigma^{-4}) = 6K_4K_2^{-2}t^{-1}+O(t^{-2}).\end{aligned}\right\} \tag{2.6}$$

The result for λ_3 is, in effect, given by Aris (1956), and Sayre (1968) makes the explicit point that (1.1) does not describe the behaviour of λ_3 correctly.

Now the K_n are only exceptionally zero. It therefore follows from (2.5) that (1.1) does not predict the correct behaviour of $\lambda_n (n \geqslant 3)$ as $t\rightarrow\infty$ (since (1.1) is obtained from (2.1) only by setting K_n zero $(n \geqslant 3)$). Since the $\lambda_n (n \geqslant 3)$ define the deviations of the observed distribution from Gaussianity it follows that there is no more justification for describing non-Gaussian distributions of C_m by non-Gaussian solutions of the heat-conduction equation than for describing them by (1.4). The only qualification, to be discussed further in §3, occurs when the initial distribution of C_m, which affects the term of $O(\sigma^{-n})$ in (2.5), is such that the term in (2.5) of $O(\sigma^{2-n})$ only dominates that of $O(\sigma^{-n})$ when both are experimentally indetectable. This only occurs when one or more cumulants are initially very large or, equivalently, when the cloud of solute is initially spread out over a distance very large compared with the mean radius of the tube cross-section.

The set of cumulants determine C_m uniquely and it is possible to derive an expression for C_m in terms of its cumulants (Chatwin 1970).

3. Some long-term effects of the initial distribution

The initial distribution affects the term of $O(\sigma^{-n})$ in (2.5), but not the leading term. The precise dependence of this term on the initial distribution can be determined, at least in theory, by forward integration of the exact equations from $t = 0$. Nevertheless, it is possible to see by means of an example that the initial distribution may strongly affect the time after which a particular λ_n is given, to good approximation, by the first term in (2.5). Indeed this time may be so large that after it has elapsed the value of the particular λ_n will be smaller than can be detected in an experiment.

Consider a single blob of solute released in a tube in which a is a typical length of the cross-section and D is a typical lateral diffusivity. If the initial variance of the blob is not much larger than a^2, then after a time of order $a^2/5D$ the dispersion process will be governed by (2.1) and after a time of order a^2/D by (1.1) (Chatwin 1970). After a total time, say T, depending on the sensitivity of the apparatus being used to measure concentration the profile will be Gaussian, with variance σ_*^2, where

$$\sigma_*^2 = 2K_2 t + \text{constant}. \tag{3.1}$$

Notice that by suitable choice of the origin of time, the constant in (3.1) can be made zero (in accordance with remarks in § 1) but this choice will for the moment be postponed.

Now consider a second experiment in which two blobs, both equal to the blob just considered, are released at a distance $2x_*$ apart. After time T the distribution of concentration in each blob will be Gaussian so that, by superposition and in accordance with (1.3),

$$C_m = \frac{1}{2\sigma_*(2\pi)^{\frac{1}{2}}}\left[\exp\left\{-\frac{(x-Ut-x_*)^2}{2\sigma_*^2}\right\}+\exp\left\{-\frac{(x-Ut+x_*)^2}{2\sigma_*^2}\right\}\right]. \tag{3.2}$$

The value of x_g given by (3.2) is consistent with (1.2); the value of σ^2 is, on integration, given by

$$\sigma^2 = \sigma_*^2 + x_*^2. \tag{3.3}$$

This can be made consistent with (1.2) by virtue of (3.1) and the remarks which follow it.

The values of the absolute cumulants associated with any one blob cannot be detected, by definition of T. However, it does not follow that the same is true for the distribution formed by both blobs since the absolute cumulants are not linear functions of C_m. Clearly the odd cumulants are zero by symmetry but, for example,

$$\lambda_4 = -2(x_*/\sigma)^4 = -\tfrac{1}{2}(x_*^2/K_2 t)^2 \tag{3.4}$$

on integration. This is consistent with (2.5) since the term involving K_4 became negligible after time T. Thus if $x_*^2 \gg K_2 T$ there will be a substantial period during which λ_4 is significant and determined solely by the initial distribution. After a time much greater than x_*^2/K_2, the value of λ_4 and higher cumulants will be negligible and the form (3.2) will reduce to the single Gaussian expression, (1.4).

The example just given does not invalidate the result of § 2, that the development of the higher-order cumulants cannot be described by (1.1) as $t \to \infty$, but shows rather that the cumulants may be negligibly small before (2.5) becomes useful, if the initial variance of the *whole* distribution is much larger than a^2.

I am indebted to Professor G. K. Batchelor for suggesting the useful example just discussed.

REFERENCES

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

CHATWIN, P. C. 1970 *J. Fluid Mech.* **43**, 321.

KENDALL, M. G. & STUART, A. 1958 *The Advanced Theory of Statistics*, vol. 1. London: Griffin.

SAYRE, W. W. 1968 *J. Hyd. Div., Proc. A.S.C.E.* **94**, 1549.

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

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