VARIABILITY AND HEAVY GAS DISPERSION

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Summary

Recent experimental work on heavy gas dispersion, confirming that the degree of variability between apparently identical releases is not small, is discussed in terms of a simple statistical framework. It is concluded (consistent with earlier work) that there is strong justification for the further development of mathematical models for hazard assessment that take explicit account of variability. Such models will, of course, have to be tested experimentally but it is argued that, in practice, experimental estimates of the degree of variability depend strongly on the detailed characteristics of the concentration sensors, and some model calculations are presented in support of this view. The role of molecular diffusion is also considered as is, finally, the question of the number of repetitions of a release that are needed to obtain reliable estimates of statistical properties.

Introduction

This paper is natural successor to an earlier paper [1] on the same topic. However some material from that earlier paper will be repeated here so that the present paper can be self-contained.

The evolution in time and space of two separate releases of heavy gas into the atmosphere (and indeed of any trials involving turbulence) can never be the same, however much care is taken in the attempt to have conditions identical immediately before release. Such conditions will include the ambient atmospheric turbulence, and the size, shape and density of the initial cloud. This paper and the earlier one deal with the differences between separate releases (here termed variability), and the quantitative description and practical importance of such differences.

Clearly the degree of variability depends on how closely conditions before release are controlled. This point is crucial in an understanding of variability. To illustrate its importance, consider a series of trials in each of which all characteristics of the cloud of heavy gas immediately before release are identical. Now suppose first that all trials in the series occur under neutrally stable conditions with the wind at release in the same direction and having

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the same (mean) speed. While there will be variability between trials in this series, its degree will obviously be less than that in a second series in which atmospheric conditions at release are arbitrary. Variability in this second series will be affected by initial differences in wind speed and direction, and in atmospheric stability, factors which are deliberately excluded from the first series.

Quantitative assessment of variability is therefore sensible, and indeed possible, only if the series of releases to which the variability relates is precisely defined. The technical term for such a precisely defined series is "ensemble", a concept rightly called fundamental by Lumley and Panofsky [2, p. 6]. As noted in [1] and elsewhere, a choice of an underlying ensemble is compulsory before every analysis of a phenomenon involving turbulent dispersion. In most cases this choice will be made on practical grounds; thus for the assessment of hazards associated with heavy gas clouds, the ensemble is likely to be one that includes (in proper proportions for the location) all weather conditions. It should be noted, however, that it is perfectly valid to regard this ensemble as the union of many "sub-ensembles", each representing release under specific weather conditions; when this point of view is adopted, there remains, of course, the non-trivial problem of combining the separate estimates from each sub-ensemble to form an estimate representative of the whole ensemble.

Failure in the past to specify the choice of ensemble in explicit terms has led to much confusing controversy, almost all of it unnecessary.

Whatever the underlying ensemble, the inevitable turbulence ensures that the concentration $\Gamma(\underset{\sim}{x}, t)$ of the dispersing gas in any one release (or "realization") of the ensemble is unpredictable, i.e. it is a random variable for each position $\underset{\sim}{x}$ and time after release t. Throughout this paper $\Gamma(\underset{\sim}{x}, t)$ is always a concentration defined according to the continuum hypothesis [1], but the particular units in which $\Gamma(\underset{\sim}{x}, t)$ is measured (e.g. volume ratio or mass per unit volume) are of little importance. In view of a comment made at the Symposium in the discussion following this paper, it is also appropriate here to clarify an apparent misunderstanding regarding the definition of $\underset{\sim}{x}$, specifically the position for which $\underset{\sim}{x} = \underset{\sim}{0}$. This position is a matter of choice; the many possibilities include:

 (i) $\underset{\sim}{x} = \underset{\sim}{0}$ is, for all t, the centre of mass of the cloud at release, i.e. a position fixed in space;

 (ii) $\underset{\sim}{x} = \underset{\sim}{0}$ is, for each t, the instantaneous centre of mass of the cloud, i.e. a position randomly moving in space;

 (iii) $\underset{\sim}{x} = \underset{\sim}{0}$ is, for each t, the position of a point moving with the mean wind and coincident at $t = 0$ with the centre of mass of the cloud at release, i.e. a position moving deterministically in space.

Possibilities (i) and (ii) are those normally used in "absolute diffusion" and "relative diffusion" respectively, and, in effect, each of the three possibilities yields a separate ensemble. In line with earlier comments, it is evident (for example) that the variability when possibility (i) is adopted is much greater than that with possibility (ii), because with relative diffusion the large varia-

bility due to random meandering of the cloud as a whole is totally eliminated. From the point of view of practical needs in hazard assessment, only possibility (i) seems ultimately viable because results and predictions are needed with respect to fixed spatial locations.

Given that $\Gamma(\underset{\sim}{x}, t)$ is a random variable, there exists a probability density function (or p.d.f.) of concentration $p(\theta; \underset{\sim}{x}, t)$ with the property that $p(\theta; \underset{\sim}{x}, t)\delta\theta$ is the probability in a single realization of the ensemble that the actual concentration $\Gamma(\underset{\sim}{x}, t)$ lies between θ and $\theta + \delta\theta$, where $\delta\theta$ is small and positive. The (ensemble mean concentration $C(\underset{\sim}{x}, t)$ and the mean square fluctuation $\overline{c^2}(\underset{\sim}{x}, t)$ are the mean and variance respectively of the distribution whose p.d.f. is $p(\theta; \underset{\sim}{x}, t)$; thus, following standard statistical definitions*,

$$C(\underset{\sim}{x},t)=\int_0^\infty \theta \, p(\theta;\underset{\sim}{x},t)\mathrm{d}\theta \quad \overline{c^2}(\underset{\sim}{x},t)=\int_0^\infty \{\theta-C(\underset{\sim}{x},t)\}^2 \, p(\theta;\underset{\sim}{x},t)\mathrm{d}\theta \tag{1}$$

As explained in detail in [1], C and $\overline{c^2}$ will normally be estimated experimentally by appropriate arithmetical averages of the results of many realizations. Note that the common technique that uses time averages is legitimate only when conditions are statistically stationary (so that p, C and $\overline{c^2}$ are independent of t), and this was not so at Thorney Island.

When variability exists, $\overline{c^2}(\underset{\sim}{x},t)$ is non-zero and *vice versa*. Thus one natural measure of variability is $I(\underset{\sim}{x},t)$, where

$$I(\underset{\sim}{x},t) = \frac{\sqrt{\overline{c^2}(\underset{\sim}{x},t)}}{C(\underset{\sim}{x},t)}. \tag{2}$$

Although $I(\underset{\sim}{x},t)$ is probably the easiest measure to determine reliably from experiments, there are others that merit consideration. In particular, there is $p(\theta; \underset{\sim}{x}, t)$ itself, and there is the peak-to-mean ratio.

It is worthwhile to insert here some remarks about the latter measure. The peak concentration measured in experiments is a highly random variable, as shown for example in Figure 15 of Koopman et al. [3]. This is easily understandable since actual concentration distributions are highly intermittent, and have large spatial and temporal gradients; thus, as a consequence of the inevitable instrument smoothing, measured peak concentrations are normally much less, and by an unpredictable amount, than θ_{max}, the actual peak concentration. It therefore does not seem sensible to base predictive models for hazard assessment (or anything else!) on measured peak-to-mean ratios. Even from a theoretical point of view, the value of θ_{max} can have little real significance since (almost by definition) the probability of actual concentrations being near θ_{max} is very small.

In all ways $I(\underset{\sim}{x},t)$ — or, equivalently, $C(\underset{\sim}{x},t)$ and $\overline{c^2}(\underset{\sim}{x},t)$ — is a much more satisfactory measure of variability than the peak-to-mean ratio. So, more ambitiously, is $p(\theta;\underset{\sim}{x},t)$ and the earlier paper [1] contains a summary of

*The upper limit in the integrals in (1) is taken as ∞ so that the definitions are valid for all choices of the units of concentration. When the concentration is defined as a volume ratio, $p(\theta; \underset{\sim}{x}, t)$ is identically zero for $\theta > \theta_{max}$ (where $\theta_{max} \leqslant 1$ is the maximum concentration) so that the upper limits can be replaced by θ_{max}.

methods for hazard assessment based on it — methods whose validity has been illustrated in experiments by Birch, Brown and Dodson [4].

Recent experimental evidence

Deterministic models for hazard assessment ignore variability and therefore (in effect) assume that $I(\underset{\sim}{x},t) \ll 1$. However, evidence summarized in [1] indicates that $I(\underset{\sim}{x},t)$ is never very small but takes values of order unity and greater. The experimental part of this evidence relied on data taken in statistically steady flows, and it is therefore important to consider two sets of measurements (taken since [1] was written) on fluctuations associated with statistically unsteady dense gas clouds.

Hall et al. [5] presented wind tunnel data taken in experiments designed to simulate some of the heavy gas trials on Porton Down [6, 7]. For some of these trials, three repeat simulations were performed, each with the same release conditions (although, of course, the naturally occurring turbulent fluctuations in the ambient wind could not be controlled). Table 1 gives some details, while Figs. 1 and 2 show typical results of the graphs of measured concentration against time since release obtained in these repeat simulations.

In an extensive series of wind tunnel tests using Freon-12 (CCl_2F_2) for which $\rho_g/\rho_a = 4.2$, Meroney and Lohmeyer [8] measured concentrations in a wind tunnel for a variety of release conditions. Later [9] they considered the magnitudes of the measured concentration fluctuations, and Fig. 3 shows, for a near ground level sensor, the derived values of I plotted against non-dimensional downstream distance. The scatter in Fig. 3 is large partly because most of the points are the averages of five replications only; obviously many more replications are needed to achieve stable estimates.

Another probable cause of the scatter is that the values of Ri_0 (defined in Table 1) vary for points in this figure, although all values exceed about 450.

TABLE 1

Some details of two Porton trials (numbers approximate only)

Figure No. in this paper	Porton trial No.	u_* (m s^{-1})	$\dfrac{\rho_g - \rho_a}{\rho_a}$	Ri_0	Sensor position relative to release
1	21	0.41	0.3	60	Ground level; 25 m downwind.
2	33	0.15	1.2	1800	Ground level; 15 m dowwind.

Notation used. ρ_a = air density; ρ_g = initial gas density; u_* = shear velocity in undisturbed atmosphere; $Ri_0 = g(\rho_g - \rho_a)H_0/\rho_a u_*^2$ = initial Richardson number; H_0 = initial cloud height.

High values of Ri_0 cause the dispersion soon after release to be dominated by the cloud's negative buoyancy and not by the ambient atmosphere. Thus the values of I soon after release seem likely to decrease as Ri_0 increases [10]; since Ri_0 is an increasing function of $H_0 \sim V_0^{1/3} \sim L_0$, the two pairs of curves marked on Fig. 3 are consistent with this belief.

It is clear from Figs. 1 to 3 that, in general, variability is not small and, consistent with the earlier evidence reported in [1], that values of I of order unity are typical. Hall et al. [5, p.30] make the following comment: ". . . while the model and full-scale concentration measurements sometimes show significant differences there are a number of plausible reasons for this, particularly the high levels of variability in repeat runs that can occur in the
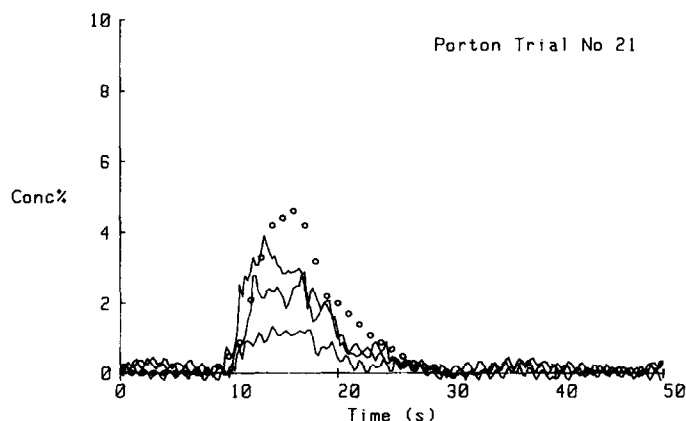

Fig. 1. A wind tunnel simulation of Porton trial No. 21 by Hall et al. [5]. The solid lines indicate the results of the three repeat simulations and the open circles are the results in the trial. Data details are given in Table 1.
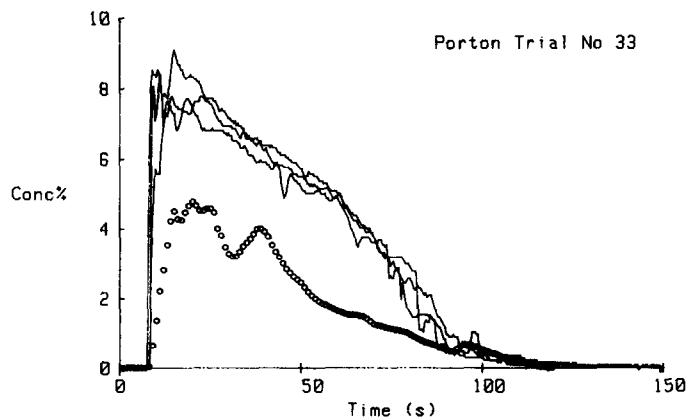

Fig. 2. A wind tunnel simulation of Porton trial No. 33 by Hall et al. [5]. Refer to Table 1 and the caption to Fig. 1.
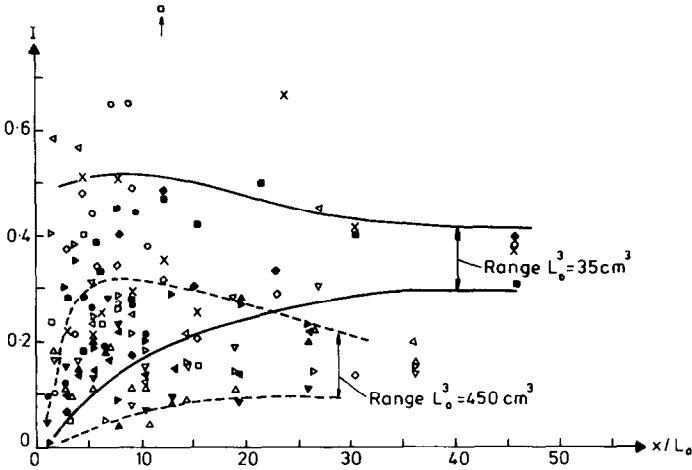
Fig. 3. Wind tunnel measurements of I by Meroney and Lohmeyer [8, 9]. Here x is distance downstream and the different symbols represent different release volumes and wind speeds, with each being the result of at least five repetitions. Details are given in Table 8 of [8] and Figure 13 of [9]. It should also be noted that the values of I are derived from fluctuations in the maximum recorded concentration at position x and therefore, strictly speaking, are not the same as the values considered in the text. However, it is difficult to see that the difference is important for present purposes.

model and would exist at the full-scale. There is no way of resolving these differences and the conclusion of the comparison must be that the agreement between model and full-scale measurements is the best that is likely to be achieved where an attempt at assessing effects due to variability has not been performed for both sides of the comparison. Hopefully this aspect will be rectified in any future exercise". In similar vein, Meroney and Lohmeyer [8, p.152] recommend: "The statistics of a single cloud release situation should be measured in the laboratory in detail. Even including measurements of this report, such information is scarce."

Such observations, and others made during the Sheffield Symposium, seem to indicate a movement since [1] was written towards general acceptance of the main thesis of that paper, namely that practical models of hazard assessment that explicitly incorporate variability ought to be developed and tested.

Some theoretical considerations

It is helpful now to proceed by considering an idealized situation. Suppose that the ensemble is such that at release at $t=0$ the cloud of heavy gas has the same shape, the same volume L_0^3, and the same uniform concentration θ_0 in each realization of the ensemble. For the time being, make the additional assumption that molecular diffusion has negligible effect. This assumption,

while certainly not valid for the whole history of the dispersion, is very useful in a preliminary analysis because it helps understanding. The philosophy is that of Batchelor [11] and the early part of Chatwin and Sullivan [12].

Under these conditions the actual concentration $\Gamma(x,t)$ must have one of only two values: θ_0 and 0. It follows that $p(\theta;x,t)$, the p.d.f. of concentration introduced earlier, must have the form [13]

$$p(\theta;x,t)=\pi(x,t)\delta(\theta-\theta_0)+[1-\pi(x,t)]\delta(\theta), \tag{3}$$

where $\pi(x,t)$ — often called the intermittency — is the probability that the point x is, at time t, immersed in gas. Conversely $[1-\pi(x,t)]$ is the probability that the point x is, at time t, immersed in air. The simple form of (3), in which δ is the Dirac delta function, occurs because, when there is no molecular diffusion, every point must either lie in gas at concentration θ_0 or in air at concentration 0. A special form of (3) was given as eqn. (14) of [1].

Use of eqn. (3) in eqn. (1) gives

$$C(x,t)=\theta_0\pi(x,t); \quad \overline{c^2}(x,t)=\theta_0^2[\pi(x,t)-\pi^2(x,t)]. \tag{4}$$

Hence, from eqn. (2),

$$I(x,t) = \sqrt{\frac{1}{\pi(x,t)} - 1} = \sqrt{\frac{\theta_0}{C(x,t)} - 1}. \tag{5}$$

Some comments on these simple results are appropriate. All consequences of the random advection on $p(\theta;x,t)$ are contained in the intermittency $\pi(x,t)$. Note especially that π and therefore p are deterministic fields which depend, perhaps strongly, on the choice of ensemble (as well as on x and t). Thus, when $x = 0$ is fixed in space (absolute diffusion) π contains the effects of the meandering of the cloud as a whole, but when $x = 0$ is the instantaneous centre of mass of the cloud (relative diffusion) this meandering is excluded and only random dispersion relative to the centre influences π. In either of these cases (and in all other cases) the region occupied by gas in each realization is a randomly distorted shape of constant volume L_0^3, and this shape is randomly distributed in a region of x-space. Let the volume of this region of x-space have order of magnitude $\tilde{L}^3(t)$, where L (like π) depends on the choice of ensemble. One way, in principle at least, of estimating $L^3(t)$ would be to form a composite cloud by superposing the individual clouds from many separate realizations, such superposition being consistent with the particular ensemble, and then to take L^3 as the volume occupied by the composite cloud. It is then immediately clear, for example, that the value of L^3 for an absolute diffusion ensemble will be much greater than that for a relative diffusion ensemble at the same time after release because, in the former case, the composite cloud is likely (because of meandering) to contain many individual clouds that do not overlap, whereas, in the latter case, the choice of $x = 0$ ensures that the centres of all the individual clouds coincide. From the definition of L^3 it is now apparent that for most x (i.e.

outside any "core" there might be in the structure — see Chatwin and Sullivan [12, 14]),

$$\pi \sim \left(\frac{L_0}{L}\right)^3, \tag{6}$$

where $\sim$ denotes "order of magnitude", and therefore from eqn. (4) that

$$C \sim \theta_0 \left(\frac{L_0}{L}\right)^3 = \frac{Q}{L^3}; \quad \overline{c^2} \sim \theta_0^2 \left(\frac{L_0}{L}\right)^3 \left\{1 - \left(\frac{L_0}{L}\right)^3\right\} = \frac{Q^2}{L^3 L_0^3} \left\{1 - \left(\frac{L_0}{L}\right)^3\right\}, \tag{7}$$

where Q is the total quantity of dispersing gas in arbitrary units. In passing, note that the result for C in eqn. (4) provides a second, and more practically viable, way of estimating L^3. From these results it also follows that

$$I \sim \sqrt{\left(\frac{L}{L_0}\right)^3 - 1}, \tag{8}$$

using eqn. (2) or eqn. (5).

Evidently L^3 will normally be much greater than L_0^3 except very soon after release. Then the results above for $\overline{c^2}$ and I can be approximated by

$$\overline{c^2} \sim \frac{Q^2}{L^3 L_0^3}; \quad I = \frac{(\overline{c^2})^{1/2}}{C} \sim \left(\frac{L}{L_0}\right)^{3/2} \gg 1. \tag{9}$$

The first of these results was derived by an alternative method in [12]. For releases of ambient density gases into a neutrally stable atmosphere, L is approximately proportional* to $u_* t$, where u_* is the shear velocity [15], so eqn. (9) gives I proportional* to $(u_* t/L_0)^{3/2}$. For the buoyancy dominated stages of heavy gas dispersion, L^3 can be estimated through box model theory [16, 17] as $R^2 H$, where $R=R(t)$ and $H=H(t)$ are the radius and the height of the cylinder within which the gas is assumed to be contained. Then $I \sim \{1+(t/t_0)\}^{1/2} (H/H_0)^{1/2}$ where t_0 is the buoyancy spreading time scale (of the order of 0.5 s at Thorney Island) and H_0 is the initial cloud height. Different box models predict different variations of H/H_0 with t [18, 19] with, typically, H/H_0 first decreasing and then increasing. In any event, the effects of negative buoyancy cause the values of I for heavy gas dispersion to grow much less rapidly with t, at least until these effects become negligible so that the gas cloud behaves passively. Since the degree of negative buoyancy is measured by the initial Richardson number Ri_0, defined in Table 1, it is now clear that the physical reason for the decrease of variability with increasing Ri_0 — discussed in the previous section — is simply that gravity greatly reduces the mean cloud size $L^3(t)$.

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*From earlier remarks in this section it will be apparent that the constants of proportionality depend on the ensemble, being greater for absolute diffusion than for relative diffusion.

A puzzle and its possible causes

Use of eqns. (4) and (5) gives

$$I^2 = \frac{\overline{c^2}}{C^2} = \frac{\theta_0}{C} - 1 \approx \frac{\theta_0}{C}, \tag{10}$$

where, in view of the universally observed rapid mixing, the approximation seems likely to be valid for heavy gas clouds almost immediately after release. Recall that eqn. (10) is not merely a statement about order of magnitudes, but is exact provided the initial cloud has uniform concentration θ_0 and provided the effects of molecular diffusion are negligible*. When, as in the experiments of Meroney and Lohmeyer [8, 9], the release is of pure gas with concentrations measured as volume ratios, the value of θ_0 is 1 and so

$$I \approx \frac{1}{C^{1/2}}. \tag{11}$$

Figure 4, taken from [9], shows the data of Fig. 3, but with I now plotted against C. Although the scatter is large for the reasons already indicated, there is a general indication (noted in [9] by Meroney and Lohmeyer) that I decreases with C. However 95% of the data lie below the solid line on Fig. 4, which is a graph of

$$I = \left(\frac{0.02}{C}\right)^{1/2} \approx \frac{0.14}{C^{1/2}}. \tag{12}$$
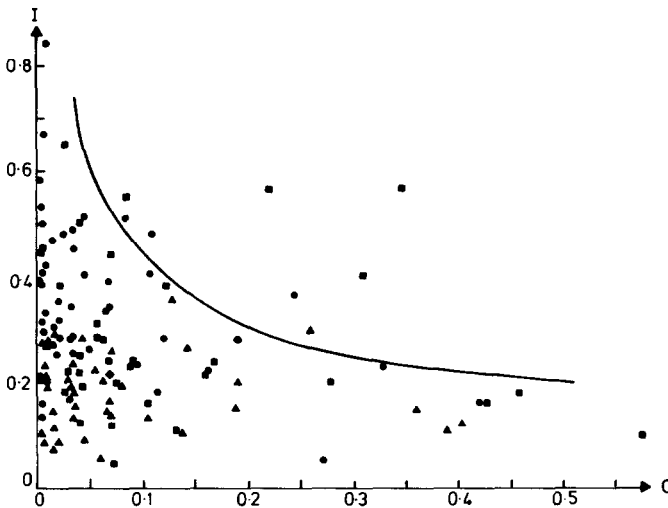

Fig. 4. Wind tunnel measurements of I by Meroney and Lohmeyer [8, 9]. Refer to the caption to Fig. 3.

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*Note that eqn. (10) holds whatever the ensemble, i.e. the differences between different ensembles is entirely due to the inevitable differences in values of $C=C(\underset{\sim}{x},t)$.

The large discrepancy between the constants in eqns. (11) and (12) needs and merits further investigation. There are only three possible causes of the discrepancy. These are:
(1) the initial cloud is not of uniform concentration;
(2) the effects of molecular diffusion are important;
(3) the large difference is due to instrumentation effects.

The effect of (1) alone would, in general, be to give a range of actual concentrations, hence causing the actual p.d.f. to be much more complicated than the simple form in eqn. (3). However this effect was not present in the pure gas releases from which the data in Fig. 4 were derived and cannot therefore explain the discrepancy between eqns. (11) and (12). Nor does this effect seem likely to be significant in any analysis of variability in the Thorney Island trials since D.R. Johnson of N.M.I. Ltd. informs us that very little departure from uniformity was observed during the monitoring of gas concentrations immediately prior to release. More generally, this effect by itself cannot cause order of magnitude departures from eqn. (11) since the order of magnitude estimates of C and $\overline{c^2}$ in eqns. (7) and (9) respectively hold [12] irrespective of whether the p.d.f. is given by eqn. (3).

The conclusion therefore is that the resolution of the puzzle must lie in effects (2) and/or (3). It is helpful first to consider effect (3) in isolation, thereby postponing the discussion of molecular diffusion until later.

Instrument smoothing

The potential importance of instrument smoothing was discussed in [1] and it is useful to summarize the main points made there. Casual observation of any single realization of a turbulent diffusion phenomenon, e.g. smoke from a cigarette, shows a very fine-scale spatial structure in the concentration field which, because of the advection, is inevitably accompanied by very rapid temporal changes. According to estimates in [1], typical length and time scales for this fine structure are of order 10^{-3} m and 10^{-3} s respectively in field trials like those at Thorney Island. Although many different types of instrument are used to measure gas concentrations it is fair to say that none is capable of resolving simultaneously both the fine scale spatial structure (in each of three dimensions) and the rapidly changing temporal structure.

The degree of instrument smoothing has been estimated for various types of sensor commonly used in wind tunnels. The data shown in Figs. 1 to 4 were obtained using aspirated hot-wire anemometers of a type whose response time is of order 10^{-3} s, according to Wilson and Netterville [20]. Calculations by Perry [21] suggest that the response time is even smaller (down to order 10^{-5} s) for a probe using the same principle but of the type described, for example, by Brown and Rebollo [22]. However the spatial resolution of such instruments seems less satisfactory since measurements involve averaging over a spatial volume in the approximate shape of a cylinder of radius of order 2×10^{-3} m and length of order several millimetres. For the

data in Figs. 1 and 2, for example, Hall et al. [5] state that the spatial resolution was typically of a few millimetres. According to Fackrell [23], similar response times and spatial resolution occur with the flame ionisation detector used to obtain the data in Fackrell and Robins [24, 25] quoted in [1].

Instruments used in field trials, such as the ion generator-collector system of Jones [26] and the oxygen deficiency meters used at Thorney Island, are normally larger than laboratory probes since, necessarily, they are much more robust. Although much improvement has occurred in reducing response times, the spatial resolution achieved in field trials, including Thorney Island, is typically of order centimetres rather than millimetres.

The need to consider instrument smoothing in interpreting the results of turbulent diffusion experiments has, of course, long been recognized. Most accounts, such as that in Pasquill [27, pp. 11—22], are almost exclusively in terms of Fourier spectra. For phenomena that are intrinsically unsteady and inhomogeneous (in a statistical sense), like the Thorney Island trials, use of Fourier spectra to present the data has little merit since comparison with theory is not effectively made easier, yet the data are taken one step further away from reality. Because of the wide variety of instruments that are used in practice, it is obvious that no simple rule will be adequate in all cases where smoothing is important. At present, it is realistic only to consider simple models with the aim simply of assessing whether smoothing is likely to be significant.

Here particular concern will be with modelling the effects of finite spatial resolution on C, $\overline{c^2}$ and I, using the idealized situation shown schematically in Fig. 5, which represents a one-dimensional traverse across the interior of a cloud. The concentration is non-zero, with value θ_0, only in blocks of constant thickness λ, and the distance X between successive pairs of blocks is a random variable with p.d.f. $f(\xi)$, where $0 \leqslant \xi < \infty$ and $f(\xi)$ is the same for all pairs of blocks. Suppose that the mean of X is η, so that

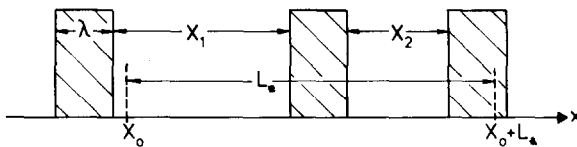$$\eta = \int_0^\infty \xi f(\xi) \mathrm{d}\xi. \tag{13}$$

Fig. 5. Definition sketch for a one-dimensional model of instrument smoothing. Note that X_1 and X_2 are two random values of X.

This model is one for which the p.d.f. of concentration $p(\theta;\underset{\sim}{x},t)$ is given by eqn. (3) with, clearly, the intermittency given by

$$\pi(\underset{\sim}{x},t) = \pi_0 = \frac{\lambda}{\eta + \lambda}. \tag{14}$$

Thus, using eqn. (4),

$$C = \pi_0 \theta_0; \quad \overline{c^2} = (\pi_0 - \pi_0^2)\theta_0^2. \tag{15}$$

Now suppose that this distribution of concentration is sampled with a probe of width L_* (and zero time constant) so that the measured concentration Γ_m is

$$\Gamma_m = \frac{1}{L_*} \int_{X_0}^{X_0 + L_*} \Gamma(x)dx, \tag{16}$$

where X_0 is uniformly distributed (so that π_0 in eqn. (14) is the probability that X_0 is in one of the blocks of non-zero concentration), and $\Gamma(x)$ is the actual concentration, equal to θ_0 or 0. It follows immediately from eqn. (16) that C_m, the ensemble mean measured concentration, satisfies

$$C_m = \frac{1}{L_*} \int_{X_0}^{X_0 + L_*} Cdx = C, \tag{17}$$

since the random variables X_0 and Γ are independent; thus the mean concentration is unaffected by smoothing. On the other hand $\overline{c_m^2}$, the ensemble mean square measured fluctuation, is changed according to the well-known formula [e.g. 2, p. 36]

$$\frac{\overline{c_m^2}}{\overline{c^2}} = \frac{2}{L_*} \int_0^{L_*} \left(1 - \frac{x}{L_*}\right)\rho(x)dx, \tag{18}$$

where

$$\rho(x) = \overline{c(x+y)c(y)}/\overline{c^2}, \tag{19}$$

is the spatial autocorrelation of the fluctuating concentration. Determination of the form of $\rho(x)$ requires specification of $f(\xi)$; the obvious choice, consistent with eqn. (13), is

$$f(\xi) = \eta\, e^{-\xi/\eta}. \tag{20}$$

Use in eqn. (18) of the $\rho(x)$ consistent with eqn. (20) leads, after calculations whose details will be given elsewhere, to values of $\overline{c_m^2}/\overline{c^2}$ which are typified by the graphs in Figs. 6. Use of eqn. (18), but in models of the effects of time averaging, has also been made by Venkatram [28] and by Sykes [29].

The idea of this simple model is, of course, that it may qualitatively represent the intermittent structure within a gas cloud or plume so that λ is of order 10^{-3} m (i.e. λ is of the same order as the conduction cut-off length λ_c discussed in [1, 12, 14]). While observed values of the intermittency π_0 vary

greatly between zero and unity, as shown by the results of Jones [26] and Fackrell and Robins [24, 25], Fig. 6(a) shows greatly reduced values of the perceived mean square fluctuation for all values of π_0 even for low values of L_*/λ (relatively little spatial smoothing). Figure 6(b) shows that this trend continues, albeit at a slower rate, as L_*/λ increases for small values of π_0. Thus this simple one-dimensional model of smoothing effects predicts mea-
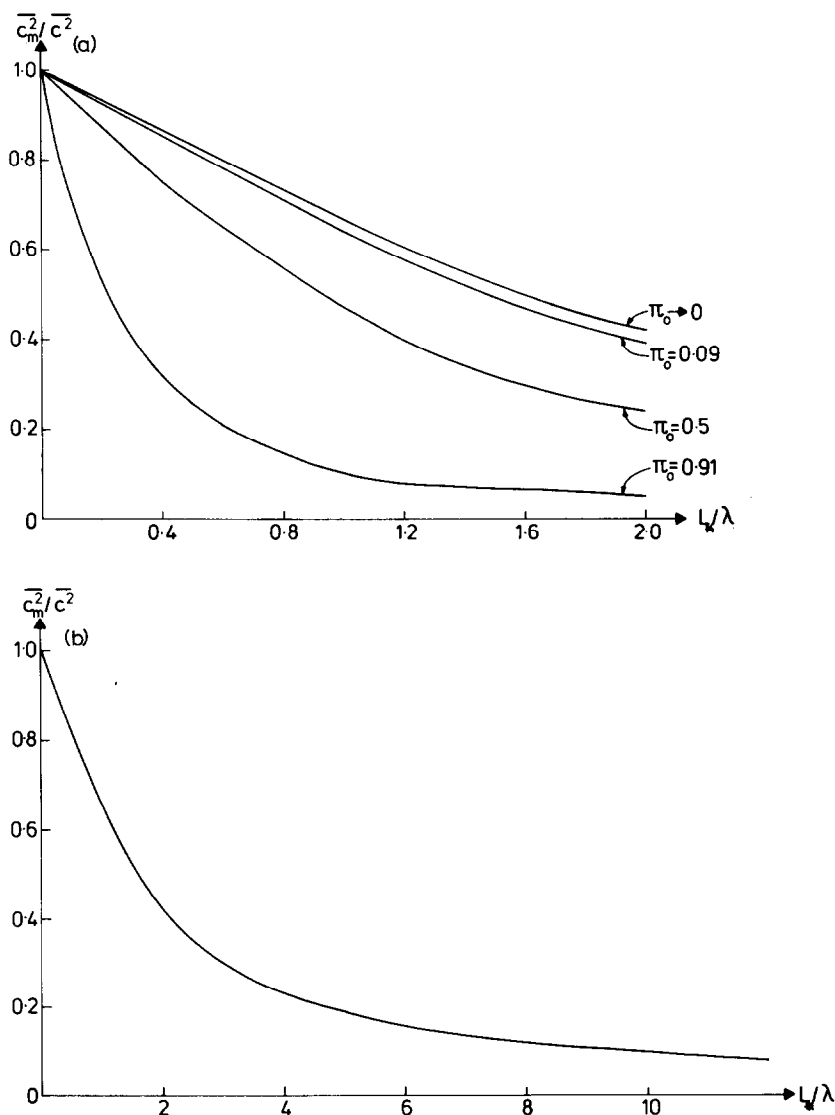

Fig. 6. (a) Model calculations of $\overline{c_m^2}/\overline{c^2}$ for various values of π_0 for $L_*/\lambda < 2$. (b) Model calculations of $\overline{c_m^2}/\overline{c^2}$ for $\pi_0 \to 0$ for $L_*/\lambda < 12$.

sured values of the mean square fluctuation that are lower by an order of magnitude than the actual values when $L_*/\lambda \sim 10$. This degree of smoothing is likely to be present in many experiments, including the Thorney Island trials.

Of course, the quantitative results in Fig. 6 cannot be applied directly to real clouds, where a traverse would give gas-containing blocks of varying widths and varying distributions of concentration. Also, because of molecular diffusion, the spaces between the blocks are likely to contain not pure air but gas at low concentrations, and geometrical differences between one- and three-dimensional space cause changes in the values of the intermittency. Real instruments give measurements affected by temporal, as well as spatial, smoothing. Finally the calculation above focuses on structure within the instantaneous cloud and takes no account of meandering, in complete contrast to the model in [29]; it is therefore primarily relevant to a relative diffusion ensemble.

Some comments on molecular diffusion

While the net quantitative effect of the complications discussed in the previous paragraph is impossible to assess, it is difficult to escape the conclusion that instrument smoothing could cause, even in well-conducted trials in wind tunnels, differences between theory and experiment that are at least of the magnitude of the difference between eqns. (11) and (12).

On the other hand, the values of $\overline{c^2}$ predicted by the idealized theory above are incorrect because of dissipation due to molecular diffusion. It is easy to derive the exact equation [see e.g. 12]

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\int C^2 \mathrm{d}V + \int \overline{c^2}\mathrm{d}V\right\} = -2\kappa\left\{\int(\nabla C)^2\mathrm{d}V + \int\overline{(\nabla c)^2}\mathrm{d}V\right\}, \tag{21}$$

where κ is the molecular diffusivity and the integrals are over the whole of space*. By mass conservation $C \sim Q/L^3$ — eqn. (7) — whether or not molecular diffusion effects are important, and so the integrals involving C in eqn. (21) both tend to zero. If the rate of this convergence to zero is sufficiently fast, eqn. (21) becomes

$$\frac{\mathrm{d}}{\mathrm{d}t}\int \overline{c^2}\,\mathrm{d}V \approx -2\kappa\int\overline{(\nabla c)^2}\mathrm{d}V, \tag{22}$$

and this equation should have (at least) qualitative validity in other circumstances. The right-hand side of eqn. (22) is negative definite and hence

$$\int \overline{c^2}\,\mathrm{d}V \to 0 \text{ as } t \to \infty. \tag{23}$$

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*In practice, of course, the integrals need only be taken over the volume over which C and $\overline{c^2}$ are effectively different from zero.

Most research on the rate of decay of $\overline{c^2}$ has occurred for conditions which are statistically steady and homogeneous, at least to good approximation, and papers by Lundgren [30] and Newman et al. [31] illustrate the two main, and contrasting, approaches to the problem. Unfortunately, the methods discussed in such papers cannot be applied easily to the normal case (and that of present interest) where conditions are neither steady nor homogeneous and, consequently, research in this area is currently rather controversial, although there is one point that is generally agreed. This is that the rate of decay of $\overline{c^2}$ depends on the release conditions, particularly L_0.

To summarize, molecular diffusion ensures that c^2 tends to zero as t increases more quickly than is given in eqn. (9). On the other hand, $\overline{c_m^2}$, the measured value, is lower, probably much lower, than $\overline{c^2}$ as the result of unavoidable smoothing. The interpretation of experimental data on variability is therefore extremely difficult. Nor has theoretical work progressed very far; Durbin [32] presented an imaginative model in which the quantity considered was a smeared value of $\overline{c^2}$, intended to represent the net effects of both molecular diffusion and instrumentation. The model does not appear to be applicable to dispersion of a cloud in three dimensions, nor does it appear likely that the two separate effects can be adequately treated in the same simple way because, for one thing, the effects of instrumentation are obviously dependent on the size of the instrument.

At present therefore the way in which these difficulties should be resolved is unknown. Moreover, as was evident from discussion at the end of the presentation of this paper at the Symposium, their consideration often leads to heated argument. Here we wish only to reiterate the views we expressed at that time. Firstly, even though molecular diffusion acts directly only on the very fine-scale structure of the concentration field, its net effect on $\overline{c^2}$ is ultimately substantial and cannot be ignored*. Secondly, we believe that measured profiles of $\overline{c^2}$ are significantly affected by instrumentation.

The assessment of variables from experiments

In experiments like those at Thorney Island (and earlier at Porton), cost prohibits the systematic repetition of a release. Thus the mean concentration $C(\underline{x},t)$ and the mean square fluctuation $\overline{c^2}(\underline{x},t)$ cannot be estimated in the standard way using arithmetical averages and discussed in [1]. It is nevertheless important to examine the results of such field trials to assess, if possible, whether the data from large-scale releases are consistent with values of C and $\overline{c^2}$ that are observed in wind tunnel experiments, and also to validate scaling laws. Such an examination of the Thorney Island data is being undertaken

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*We record also our opinion that a reliable estimate of this net effect of κ on $\overline{c^2}$ will require consideration of (a) the fact that dispersion takes place in three dimensions and (b) that there is a finite volume of dispersing gas. Consequently we are doubtful of the worth of some estimates obtained, in effect, by extending results from one-dimensional models with homogeneous conditions.

by one of us in collaboration with A. Mercer of the Health and Safety Executive, and the results will be reported later.

In the meantime, however, it is probably helpful to make some comments on another problem. When variability is to be assessed from the results of wind tunnel tests, it is sensible to estimate, in advance, the number of repetitions needed to achieve estimates with some specified reliability. Having decided the ensemble, suppose N repetitions of the release are performed in each of which the concentration Γ is measured for the same x and the same time after release t. These readings of concentration are to be used to estimate the mean concentration $C=C(x,t)$ and the mean square fluctuation $\overline{c^2}=\overline{c^2}(x,t)$. (More precisely, it will be clear from comments above that the readings can be used only to estimate the mean measured concentration C_m and the mean square measured fluctuation $\overline{c_m^2}$ but this important distinction does not affect the calculations below and will not be referred to again.) Let $\Gamma^{(n)}$ be the measured value of Γ in the nth of the N releases. Then unbiased estimates of C and $\overline{c^2}$ are m and s^2, where

$$m=\frac{1}{N} \sum_{n=1}^{N} \Gamma^{(n)}; \quad s^2=\frac{1}{(N-1)} \sum_{n=1}^{N} (\Gamma^{(n)}-m)^2. \tag{24}$$

Standard results from statistical theory [33] are that, when Γ has a normal distribution,

$$t = \frac{(m-C)\sqrt{N}}{s} \tag{25}$$

is a reading from a population with Student's t distribution with $(N-1)$ degrees of freedom, and

$$Z = \frac{(N-1)s^2}{\overline{c^2}} \tag{26}$$

is a reading from a population with the χ^2 distribution with $(N-1)$ degrees of freedom. Observations of the p.d.f. of Γ such as those in [24, 34] show that, in general, the distribution of Γ is not normal. However the central limit theorem ensures that, whatever the p.d.f. of Γ, the calculations below will be valid to a high degree of approximation.

Confidence intervals for C and $\overline{c^2}$ are obtained in the normal way. Thus the interval

$$\left(m - \frac{t_\alpha s}{\sqrt{N}}, \ m + \frac{t_\alpha s}{\sqrt{N}} \right) \tag{27}$$

will contain C for $100(1-2\alpha)\%$ of the possible values of m and s, where t_α is the appropriate reading from a table of Student's t distribution with $(N-1)$ degrees of freedom [35]. Similarly the interval

$$\left(\frac{(N-1)s^2}{\chi_\alpha^2}, \frac{(N-1)s^2}{\chi_{1-\alpha}^2} \right) \tag{28}$$

will contain $\overline{c^2}$ for $100(1-2\alpha)\%$ of the possible values of s. The choice of α is of course a matter for the experimenter. It is, however, obvious that the smaller α is, the larger are the lengths of the confidence intervals in (27) and (28) for fixed N. Hence, for a confidence interval of specified length, smaller values of α yield larger values of N.

The use of confidence intervals like those above is best illustrated by one or two specific numerical examples, and here we consider only (28), the confidence interval for $\overline{c^2}$. The treatment below is very like that in [33, pp. 56—58]. Let the midpoint of the confidence interval for $\overline{c^2}$ be s_*^2, where

$$s_*^2 = \frac{(N-1)s^2}{2} \left\{ \frac{1}{\chi_\alpha^2} + \frac{1}{\chi_{1-\alpha}^2} \right\}, \tag{29}$$

from (28). It is natural to choose N so that the half-width of the confidence interval is a specified fraction β of s_*^2; thus

$$\frac{\beta(N-1)s^2}{2} \left\{ \frac{1}{\chi_\alpha^2} + \frac{1}{\chi_{1-\alpha}^2} \right\} = \frac{(N-1)s^2}{2} \left\{ \frac{1}{\chi_{1-\alpha}^2} - \frac{1}{\chi_\alpha^2} \right\} \Rightarrow \beta = \frac{\chi_\alpha^2 - \chi_{1-\alpha}^2}{\chi_\alpha^2 + \chi_{1-\alpha}^2} \tag{30}$$

For values of $N > 30$, the distribution of $\sqrt{2\chi^2}$ is approximately normal with mean $\sqrt{2N-3}$ and variance unity. This enables (30) to be solved for N in terms of u_α, where u_α is the appropriate reading from a table of the normal distribution. The result is

$$N \approx \frac{3}{2} + u_\alpha^2 \left\{ \frac{1}{\beta^2} + \frac{1}{\beta} \sqrt{\frac{1}{\beta^2} - 1} - \frac{1}{2} \right\}, \tag{31}$$

correcting an error in [33]. The meaning of this formula is that it gives the minimum value of N which ensures that the $100(1-2\alpha)\%$ confidence interval for $\overline{c^2}$ has the required precision specified by β. Table 2 gives some illustra-

TABLE 2

Calculated values of N from eqn. (31)

α	u_α	β	N
0.05	1.645	0.25	86
0.05	1.645	0.05	2164
0.025	1.96	0.25	121
0.025	1.96	0.05	3071
0.005	2.575	0.25	207
0.005	2.575	0.05	5300

tive values of N for 90%, 95% and 99% confidence intervals. Particularly noteworthy is the great increase in N caused by increased precision, which arises because the length of the confidence interval for $\overline{c^2}$ only decreases to zero as $N^{-\frac{1}{2}}$. To give a further example to reinforce this point, the value of N when $a=0.05$ (90% confidence interval) and β is as low as 0.01 is greater than 54000! It is therefore practically viable only to carry out sufficient repetitions to obtain estimates of $\overline{c^2}$ of relatively low precision. However, calculations in [33, pp. 35—37] suggest that, for values of N of order 100, higher precision estimates of C can be obtained than for $\overline{c^2}$, although in this case the value of N is rather sensitively dependent on the measured value of I (equal to s/m), where I is defined in eqn. (2).

Concluding remarks

The evidence presented in this paper reinforces the main conclusion of [1], namely that the study of variability merits the increased theoretical and practical attention that it is receiving, despite certain serious difficulties that have become more apparent. From the point of view of assessing hazards associated with dispersing heavy gas clouds, the practical purpose of this study is to facilitate the design and validation of models that include variability explicitly, and are therefore more realistic than most existing models. Although further discussion is not included here, it should be recorded that such models are being developed in a variety of fields. Apart from that whose basis was summarized in [1], Ride [36, 37] and Griffiths and Harper [38] have published work specifically directed towards toxic hazards, and Wilson [39] has proposed, and discussed in some detail, a model primarily designed for dealing with hazards arising from pipeline rupture.

Acknowledgements

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