

THE USE OF STATISTICS IN DESCRIBING AND PREDICTING THE EFFECTS OF DISPERSING GAS CLOUDS

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Summary

Nearly all existing methods of analysing hazards associated with dispersing clouds of heavy gas are based on the mean concentration, and ignore fluctuations about the mean. It is argued that, since the root mean square value of these fluctuations is not small compared with the mean, new methods should be developed which take explicit account of the fluctuations. The basis of one such method, based on the probability density functions of concentration, is summarized and some of the practical points relevant to the use of this method are discussed. The proposed method is illustrated by reference to recent experimental work in methane jets.

Introduction

This paper is concerned with certain theoretical considerations relevant to the description of the dispersion of heavy gas clouds, and to the analysis of potential hazards associated with such clouds. The emphasis will be on the use of statistical concepts and techniques. For clarity, attention will be confined throughout to the case of the instantaneous release at time $t = 0$ of a finite volume Q of a heavy gas. Continuous releases will therefore not be considered, although many of the ideas discussed in the paper can be adapted to them.

The variable of primary concern in this paper is $\Gamma(\mathbf{x}, t)$, the concentration by volume of the heavy gas, where t is time since release and the vector \mathbf{x} measures location in space with respect to a suitably specified origin. It needs emphasizing that $\Gamma(\mathbf{x}, t)$ will always denote the actual concentration in a single realization of the dispersion. By the actual concentration is meant the concentration defined according to the continuum hypothesis. Thus suppose δV is a small volume of space surrounding the point \mathbf{x} at time t , and suppose it contains a volume δV_g of pure heavy gas. Then $\Gamma(\mathbf{x}, t)$ is the value of $(\delta V_g / \delta V)$ when $\delta V^{1/3}$ is both much larger than the mean distance between gas molecules ($\sim 10^{-9}$ m) and also much less than distances characteristic of the fine-scale structure of the velocity and density fields of the continuum

($\sim 10^{-3}$ m). Now the spatial resolution that can be achieved with practical concentration sensors in field trials is inevitably much greater than 10^{-3} m. In some circumstances therefore it is necessary to distinguish the measured concentration from the actual concentration, and the suffix m will be used for this purpose. Thus $\Gamma_m(\mathbf{x}, t)$ denotes the measured concentration at position \mathbf{x} and time t in a single realization of the dispersion.

In any single realization, the heavy gas is released into an atmosphere in which the air flow is turbulent. Further turbulence is generated by the buoyancy forces caused by the density difference between the released gas and the ambient air. The predominant and distinctive feature of turbulent flow is its randomness, meaning that the fluid velocity at a particular place at a particular time cannot be predicted in practice. The inevitable consequence of the randomness for the concentration field $\Gamma(\mathbf{x}, t)$ is that it also is random. The results of two separate realizations of the dispersion will inevitably be different from one another (and different of course in an unpredictable way) whatever precautions are taken to ensure that conditions at release are the same in each realization.

Consider the series, or ensemble, of all possible realizations of the dispersion in which conditions at release, such as the container geometry and the initial density of gas in the container, are the same in each realization of the ensemble. Let the separate realizations in the ensemble be numbered from 1 to infinity. Denote by $\Gamma^{(r)}(\mathbf{x}, t)$ the concentration at position \mathbf{x} and time t in the r th realization. The ensemble mean concentration $C(\mathbf{x}, t)$ is defined by

$$C(\mathbf{x}, t) = \lim_{n \rightarrow \infty} \left[\frac{1}{n} \sum_{r=1}^n \Gamma^{(r)}(\mathbf{x}, t) \right] = \bar{\Gamma}(\mathbf{x}, t), \quad (1)$$

where the overbar notation is used as a shorthand for the limit process. In practice $C(\mathbf{x}, t)$ has to be estimated as the mean of the measured values of $\Gamma(\mathbf{x}, t)$ in a finite number N of realizations, and the larger N is, the closer is the estimated value likely to be to the real ensemble mean concentration.

Since $\Gamma(\mathbf{x}, t)$ is random, the concentration in any one realization will be different from $C(\mathbf{x}, t)$. In other words $C(\mathbf{x}, t)$ can never be the concentration in one realization, just as $3\frac{1}{2}$ can never be the result of the throw of a single dice (even though $3\frac{1}{2}$ is the ensemble mean of the possible values 1, 2, 3, 4, 5, 6). Define the fluctuation in concentration by

$$c(\mathbf{x}, t) = \Gamma(\mathbf{x}, t) - C(\mathbf{x}, t); \quad (2)$$

from eqn. (1) it follows that $\bar{c}(\mathbf{x}, t) = 0$ for each \mathbf{x} and t . By definition, the ensemble mean square fluctuation $\overline{c^2}(\mathbf{x}, t)$ satisfies

$$\overline{c^2}(\mathbf{x}, t) = \overline{[\Gamma(\mathbf{x}, t) - C(\mathbf{x}, t)]^2} = \overline{\Gamma^2}(\mathbf{x}, t) - C^2(\mathbf{x}, t). \quad (3)$$

In the language of statistics $C(\mathbf{x}, t)$ and $\overline{c^2}(\mathbf{x}, t)$ are the mean and variance of the concentration at position \mathbf{x} at time t . A measure of the degree of

variability between different realizations of the ensemble is provided by $I(x, t)$, where

$$I(x, t) = \frac{\sqrt{\overline{c^2}(x, t)}}{C(x, t)}. \quad (4)$$

In the terminology on pp. 229–230 of Csanady [1], $I(x, t)$ is the relative intensity of the concentration fluctuations. Another commonly used measure of variability is the peak to mean ratio, but this is less convenient in theoretical analysis than $I(x, t)$. Furthermore this measure has the serious practical disadvantage that it is difficult to estimate the peak concentration reliably.

From the practical point of view, it would be justified to neglect the variability between different realizations of the ensemble if $I(x, t)$ were much less than 1. If this were true then the results from a single realization would, for example, form a sufficient basis for hazard assessment. But what evidence there is strongly suggests that $I(x, t)$ is not much less than 1. There are apparently no reported measurements of $I(x, t)$ for clouds of heavy (or even neutrally buoyant) gases. However values of $I(x, t)$ of order 5–10 have been recorded in plumes of material released from a continuous source at distances from the centre line of the order of the mean plume width (Csanady [1], pp. 236–242; Fackrell and Robins [2]). Theoretical work by Chatwin and Sullivan [3, 4] is consistent with large values of $I(x, t)$ in both plumes and clouds. There is no experimental or theoretical support for the hypothesis that $I(x, t)$ is much less than 1. Despite this evidence, nearly all existing methods of analysing the hazards associated with heavy gases dispersing in the atmosphere appear to ignore concentration fluctuations, and assume that knowledge of the ensemble mean concentration $C(x, t)$ is sufficient for this purpose. Descriptions and summaries of many of these methods are given, for example, by Havens [5, 6], McQuaid [7] and Fay [8].

It is the main thesis of this paper that, because they neglect concentration fluctuations and all variability between different realizations of the same ensemble, these methods are inadequate in principle, and in practice. Accordingly the principles of another method of analysing hazards are outlined in the next section, and, in the remaining sections, some important practical questions of special relevance to the hazards associated with dispersing heavy gas clouds are discussed.

The probability density function $p(\theta, x, t)$

Consider a specified ensemble of possible realizations of the dispersion of a cloud of a specified volume of a specified heavy gas. As in eqn. (1), let $\Gamma^{(r)}(x, t)$ be the concentration at position x and time t in the r th realization. Now let θ be a possible value of $\Gamma^{(r)}(x, t)$ and let $\delta\theta$ be a small increment in θ . The proportion of the realizations for which $\Gamma^{(r)}(x, t)$ lies between θ and $\theta + \delta\theta$ obviously depends on θ , and on x and t , and will vanish as $\delta\theta$ approaches

zero. Let its value be $p(\theta; \mathbf{x}, t)\delta\theta$. In statistical language $p(\theta; \mathbf{x}, t)$ is the probability density function of concentration at position \mathbf{x} and time t . More formally

$$p(\theta; \mathbf{x}, t)\delta\theta = \text{prob.}(\theta \leq \Gamma(\mathbf{x}, t) < \theta + \delta\theta), \tag{5}$$

where prob. is an abbreviation for probability. The abbreviation p.d.f. is often used for probability density function.

There are some obvious properties that p must satisfy. Since $\Gamma(\mathbf{x}, t)$ is non-negative, $p(\theta; \mathbf{x}, t)$ is identically zero for $\theta < 0$. Also the value of $\Gamma^{(r)}(\mathbf{x}, t)$ must lie between θ and $\theta + \delta\theta$ for some θ so that, on adding the proportions in all such intervals, the sum must be 1. In other words,

$$\int_0^1 p(\theta; \mathbf{x}, t) d\theta = 1. \tag{6}$$

Every p.d.f. satisfies an equation analogous to eqn. (6). Note that when, as in this paper, $\Gamma(\mathbf{x}, t)$ is defined as a concentration by volume, it can never exceed 1. Thus the choice of the upper limit in eqn. (6) as 1 ensures complete generality. But for any given ensemble, there will be a maximum attainable concentration θ_{\max} for each \mathbf{x} and t , such that $p(\theta; \mathbf{x}, t) = 0$ for $\theta > \theta_{\max}$. Note that θ_{\max} will often be strictly less than 1, as, for example, when the gas is mixed with air before release. Thus eqn. (6) can be replaced by

$$\int_0^{\theta_{\max}} p(\theta; \mathbf{x}, t) d\theta = 1. \tag{7}$$

Except at the instant of release θ_{\max} is normally an unknown quantity which varies with \mathbf{x} and t .

The ensemble mean concentration $C(\mathbf{x}, t)$ defined in eqn. (1), and the ensemble mean square fluctuation $\overline{c^2}(\mathbf{x}, t)$ defined in eqn. (3), are related to $p(\theta; \mathbf{x}, t)$ by

$$C(\mathbf{x}, t) = \int_0^1 \theta p(\theta; \mathbf{x}, t) d\theta, \tag{8}$$

and

$$\overline{c^2}(\mathbf{x}, t) = \int_0^1 (\theta - C)^2 p(\theta; \mathbf{x}, t) d\theta. \tag{9}$$

In eqns. (8) and (9) the upper limit can of course be replaced by θ_{\max} , and other ensemble mean quantities can be defined by similar equations.

The practical value of $p(\theta; \mathbf{x}, t)$ for hazard assessment can be illustrated by considering a flammable gas. After release the gas-air mixture at position \mathbf{x}

and time t will be ignitable if the value of the concentration $\Gamma(x, t)$ lies between θ_1 and θ_2 , where θ_1 and θ_2 are characteristic of the gas, and θ_1 is the lower flammable limit (LFL). For methane the conventional LFL is 0.05. Note that here the term "ignitable at position x " will mean simply that the gas-air mixture would burn at position x in the presence of an ignition source, whether or not such burning would go right back to the source or be rapidly extinguished. It then follows from the meaning of $p(\theta; x, t)$ that the probability that the gas-air mixture is ignitable at position x and time t is

$$\text{prob}(\theta_1 \leq \Gamma(x, t) < \theta_2) = \int_{\theta_1}^{\theta_2} p(\theta; x, t) d\theta. \quad (10)$$

Likewise the value of $P(x, t)$, where

$$P(x, t) = \int_{\theta_1}^1 p(\theta; x, t) d\theta, \quad (11)$$

is the probability, or chance, that in any one realization of the ensemble the concentration at position x and time t is greater than LFL. Alternatively, it is the proportion of realizations in the whole ensemble for which this concentration is greater than θ_1 .

The equation

$$P(x, t) = \alpha, \quad (12)$$

where α is a fixed constant satisfying $0 < \alpha < 1$ is, for fixed t , a surface at every point of which there is a probability α that the concentration of gas exceeds LFL. For every point outside the surface the probability that the concentration exceeds LFL is less than α . Furthermore, for fixed t , each point in space is associated with just one value of α so that it lies on a unique surface of the family whose equation is (12). Each of these surfaces intersects the ground in a curve, or contour. Note that since the cloud will spread, on the average, as t increases, the surface $P(x, t) = \alpha$ with α fixed will change its size and shape as t increases.

There are, of course, other hazards associated with gases dispersing in the atmosphere. For flammable gases there is the hazard of complete burn-up of the cloud in the presence of an ignition source at position x and time t . Ignitability at position x and time t , as defined above, is obviously a necessary condition for complete burn-up, but whether complete burn-up follows ignition depends not only on conditions at position x and time t , but also on whether there is a continuous path of ignitable material extending right back to the source. Thus whether complete burn-up follows ignition, or not, depends on conditions at places and times other than those where ignition occurred. With toxic dispersing gases, harm occurs at position x at time t if the dosage (the time integral of the concentration at position x from release up to time t) exceeds a specified value, although for some materials the trade-off between concentration and duration of exposure is non-linear for a given level of damage.

For any specific hazard, such as ignitability or those described above, a function $P(x, t)$ can be defined as the probability that hazardous conditions exist at position x at time t . In the case of ignitability, $P(x, t)$ is simply related to $p(\theta; x, t)$ by eqn. (11), but for other hazards, like those described above, $P(x, t)$ will not be so simply obtained since its value clearly depends on conditions at points other than x and/or at times other than t . In such cases $P(x, t)$ will be related to multi-point p.d.fs of $\Gamma(x, t)$, which are like the one-point p.d.f. $p(\theta; x, t)$ but refer to the concentrations at two (or more) different positions and/or times. Summaries of the properties of such functions are given by Monin and Yaglom [9, 10], who also discuss $p(\theta; x, t)$ in more detail than is given here.

Whatever the hazard, the location, for each t , of the surfaces $p(x, t) = \alpha$, or (less ambitiously) of the contours in which they intersect the ground, would be a satisfactory, even ideal, conclusion to the hazard analysis. Safety standards would be based on the location in space, and the variation in time, of the surface, or contour, on which $P(x, t)$ was equal to a predetermined value of α . The choice of this value of α is, of course, a question for politicians to decide and one on which physical science has no bearing. But it does need stressing that (in general) safety standards cannot be based on $\alpha = 0$; it can never be absolutely guaranteed that hazardous conditions will not occur at any particular place.

Previous investigations

Prior to the publication of two pioneering papers in the same year by Lundgren [11] and Monin [12], research on the turbulent dispersion of gases was mainly devoted to the properties of the ensemble mean concentration $C(x, t)$, and to the effect on this of other important work like that described by Taylor [13], Richardson [14] and Batchelor [15]. In contrast, the years since 1967 have seen a growing research effort devoted to the investigation, by both experimental and theoretical means, of the properties of $p(\theta; x, t)$ and allied functions. With some exceptions the results of this research have not yet been applied to the analysis of hazards associated with dispersing heavy gases. However there is no doubt, in my opinion (and similar beliefs are expressed on pps. 222–223, 227–233 of Csanady [1] and by Birch, Brown and Dodson [16]), that eventually almost all of the analysis of hazards in which turbulence plays a significant role will be in accordance with the framework outlined above, i.e. based on models of $p(\theta; x, t)$ and allied functions. Quantification of the word “eventually” is, of course, difficult, and depends on estimating the rapidity both of increases in appropriate experimental results and of advances in computer technology.

Although $p(\theta; x, t)$ is more difficult to estimate than $C(x, t)$ because it contains the extra dependent variable θ , the attempt would seem to be worthwhile because $p(\theta; x, t)$ contains so much more information than $C(x, t)$.

Very few measurements of $\Gamma(x, t)$ have been made in dispersing gas clouds, mainly because of the great experimental difficulty involved in keeping

accurate track of the cloud's position. The experimental determination of $p(\theta; \mathbf{x}, t)$ would, furthermore, require a large number of separate releases of the cloud followed by direct use of the definition of $p(\theta; \mathbf{x}, t)$, given above in eqn. (5) and the immediately preceding text. The same statement is, of course, true for the other statistical properties of $\Gamma(\mathbf{x}, t)$, like the ensemble mean concentration $C(\mathbf{x}, t)$, which is defined by, and must be determined using, eqn. (1). At the moment, in fact, it seems that reasons of time and cost make the direct determination of $p(\theta; \mathbf{x}, t)$, or even $C(\mathbf{x}, t)$, an impractical proposition in gas clouds, even for experiments as extensive as those reported by Picknett [17].

A long-term aim is to determine $p(\theta; \mathbf{x}, t)$ theoretically by using the exact evolution equation for $p(\theta; \mathbf{x}, t)$. This equation, and ways of handling it, are discussed by Dopazo and O'Brien [18], Janicka, Kolbe and Kollmann [19] and Pope [20]. In view of the early stages of this field of research, however, this aim does not seem likely to be realizable in the immediate future (say 5 years). None of the work referred to considers a situation whose physics is as difficult and complicated as that controlling the dispersion of heavy gas clouds.

The difficulties of determining $p(\theta; \mathbf{x}, t)$ for heavy gas clouds, both by experiments or by theoretical means, can be resolved only by further research. In my opinion, such research should concentrate, for the moment, on understanding how the form of $p(\theta; \mathbf{x}, t)$ is determined by the basic physical processes influencing the dispersion, and unnecessarily complicated and detailed mathematics should be avoided. Some understanding has already been provided through measurements of $p(\theta; \mathbf{x}, t)$ in statistically steady flows, when $p(\theta; \mathbf{x}, t)$ becomes independent of t . In such cases $p(\theta; \mathbf{x})$ can be determined by taking many separate measurements of $\Gamma(\mathbf{x}, t)$ at position \mathbf{x} in one run, constructing a histogram, and then using the definition in eqn. (5).

Examples of $p(\theta; \mathbf{x})$ determined in essentially this way by Birch, Brown, Dodson and Thomas [21] are shown in Fig. 1. The measurements were made in a jet of natural gas injected into air at a distance $10d$ downstream from the orifice, where d is the orifice diameter, and at four separate distances r from the jet axis. In Fig. 1(a), taken on the jet axis, $p(\theta; \mathbf{x})$ has a unimodal structure with θ_{\max} being about 0.8. The fact that $p(\theta; \mathbf{x}) = 0$ in Fig. 1(a) shows that there is no chance of encountering zero values of concentration (that is pure entrained air) on the jet axis. As the measuring location moves off the axis, the chance of encountering lower concentrations increases and at $r/d = 1.3$ — Fig. 1(b) — there is a significant probability of encountering entrained air. However the form of $p(\theta; \mathbf{x})$ is still unimodal with the peak corresponding to a non-zero value of concentration. Between $r/d = 1.3$ and $r/d = 1.5$ — Fig. 1(c) — the form of $p(\theta; \mathbf{x})$ becomes bimodal with the larger of the peaks being at zero concentration. As r/d increases from 1.5 to 1.8 the bimodal structure is maintained with the ratio of the amplitudes at the two peaks increasing rapidly. For large values of r/d , greater than 1.8, the peak at the non-zero value of concentration will disappear altogether.

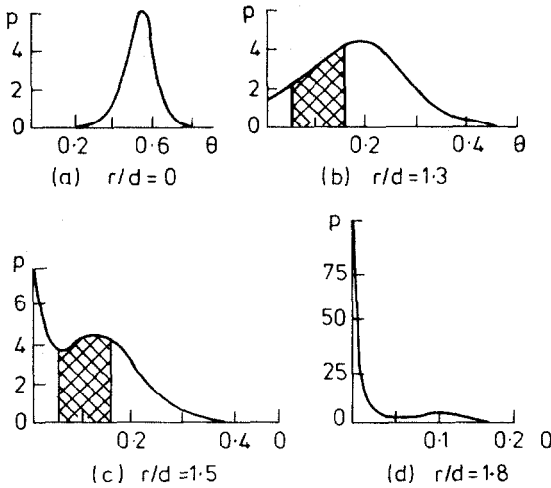


Fig. 1. Measured probability density functions of concentration by volume of methane in natural gas jet, Birch, Brown, Dodson and Thomas [21].

In accordance with eqn. (6), the area under each of the graphs in Fig. 1 is 1 (although this is not immediately obvious because of the differing scales). The hatched areas in Figs. 1(b) and 1(c) are equal to the values of $\mathcal{P}(x)$, where

$$\mathcal{P}(x) = \int_{0.05}^{0.15} p(\theta; x) d\theta \quad (13)$$

is, as stated in eqn. (10), the probability that the gas-air mixture in the jet is ignitable. In later work by the same group (Birch, Brown and Dodson [16]) the values of $\mathcal{P}(x)$ at many places in the jet were measured by generating a spark every 3 sec until either ignition was observed or until a predetermined maximum number of sparks had been generated. This procedure was repeated 400 times for each position x , and $\mathcal{P}(x)$ was taken to be the proportion of the 400 trials for which ignition (or flame formation) was observed. The experimental results are shown in Fig. 2 (as the point x at which measurements are taken moves along the jet axis) and in Fig. 3 (as x moves radially outwards in a plane at a distance $40d$ downstream from the orifice).

The experimental points were compared with theoretical curves of $\mathcal{P}(x)$, indicated by the solid lines in Figs. 2 and 3. The theoretical curves were obtained from eqn. (13) by substituting simple forms for $p(\theta; x)$, these forms being chosen on the basis of the curves shown in Fig. 1. Full details are given by Birch, Brown and Dodson [16], but, to illustrate the method, it can be noted that the solid curve in Fig. 2 was obtained by assuming that $p(\theta; x)$ was a Gaussian (or normal) distribution. This is a good approximation to the measured curve shown in Fig. 1(a). Agreement between theory and experiment is good, particularly in Fig. 2, and these experiments therefore provide

strong support for the statistical approach to hazard analysis advocated in this paper.

On the other hand, it is clear from Fig. 3 that the ensemble mean concentration $C(x)$, as shown by the dashed curve, is not, on its own, of much

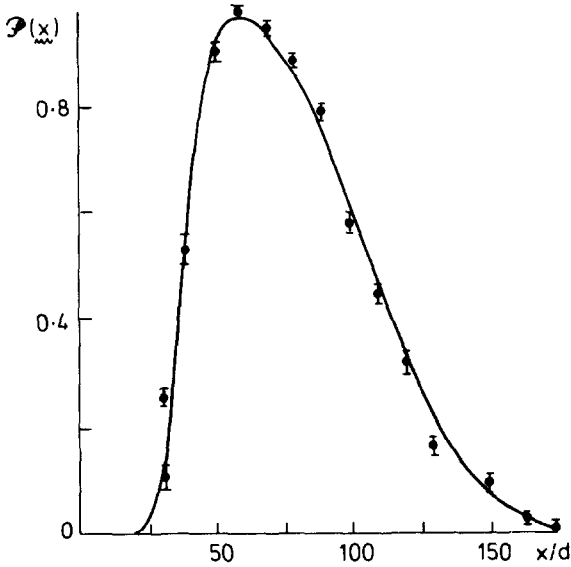


Fig. 2. Ignition probability $\mathcal{P}(x)$ along axis of natural gas jet, Birch, Brown and Dodson [16].

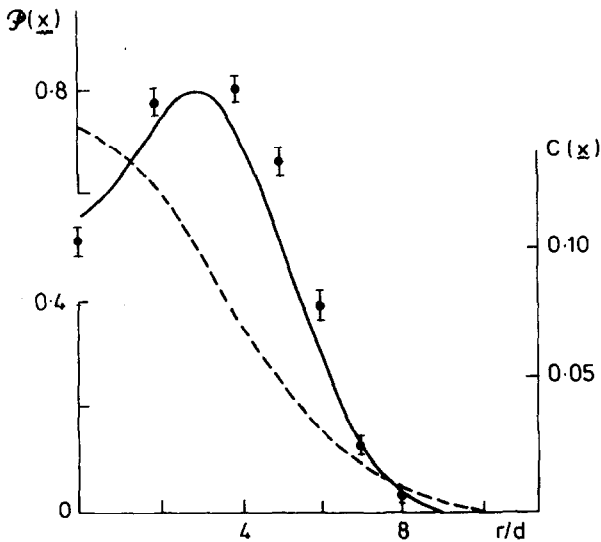


Fig. 3. Experimentally determined flammable boundary, from ref. [16] (see text).

value in estimating $\mathcal{P}(x)$. The same negative conclusion about the value of $C(x)$ as an indicator of potential hazards was reached in further experiments by Birch, Brown and Dodson [16]. In these experiments, the results of which are shown in Fig. 4, the flammable boundary for total burn-up of the jet was measured by generating a spark at a frequency of 2 kHz at each of many different points. It is clear from Fig. 4 that the measured boundary is very different from the contour on which $C(x)$ is equal to 0.05, the LFL.

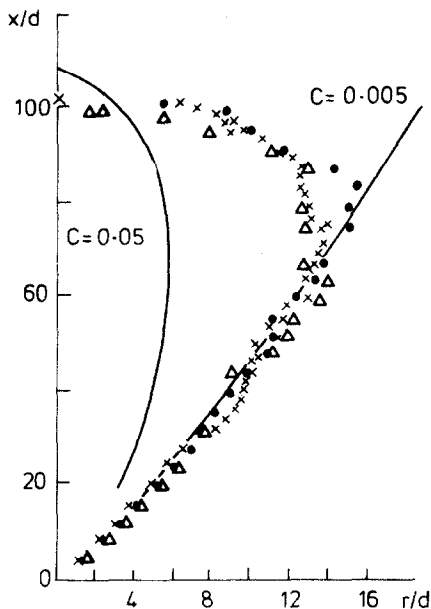


Fig. 4. Ignition probability $\mathcal{P}(x)$ in radial plane of natural gas jet, Birch, Brown and Dodson [16]. Measurements made at Reynolds numbers 12500 (Δ), 16700 (\times) and 20900 (\bullet). Solid lines are contours of ensemble mean concentration.

The concept of an ensemble, and its importance

The term “ensemble” was used earlier in the phrase “ensemble of all possible realizations of the dispersion”, but it is now appropriate to emphasize the fundamental importance of this concept for the analysis of hazards associated with dispersing heavy gas clouds (or, indeed, any phenomenon affected by turbulence). Continue to suppose that in each realization of the dispersion, a fixed quantity of gas occupying a specified shape is released by an identical mechanism at $t = 0$. Now consider two separate series of realizations, each satisfying these conditions. Suppose that in the first series no special control is exerted over the time of day, or the

ambient atmospheric turbulence (including wind speed and stability) when release occurs, and suppose that x is measured throughout each realization relative to the initial centre of mass of the cloud. On the other hand suppose that in each realization of the second series, release occurs at the same time of day under the same ambient atmospheric conditions, and that x is measured throughout each realization relative to the instantaneous centre of mass of the cloud.

Clearly (since the ambient flow is turbulent in each realization of each series) the value of $\Gamma(x, t)$ for fixed x and t will differ from realization to realization in both the first and second series. However, and this is the essential point, the statistical properties of $\Gamma(x, t)$ for the first series will be different from those for the second. It is, for example, evident that there will be far more variability from realization to realization in the first than in the second series.

The conclusion is that the statistical properties of $\Gamma(x, t)$, including $p(\theta; x, t)$, $C(x, t)$ and $c^2(x, t)$, can be meaningfully defined only for a specified series, or ensemble, of realizations. The term "mean concentration" has no meaning unless it has first been made clear what ensemble the mean is taken over.

This last point can be illustrated by considering a "box model" of the dispersing cloud (van Ulden [22]). The assumption is that at time t after release, the gas is uniformly distributed within a circular cylinder (the "box") of radius R and height H , where R and H depend on t . Then the gas concentration $\Gamma(x, t)$ is $(Q/\pi R^2 H)$ if the point x lies within the cylinder and 0 otherwise. Suppose also that the cylinder is advected horizontally by the wind in a random direction, where this direction changes from realization to realization but remains fixed during any one realization. Let this advection take place at a constant speed U . Then, at time t after release from O the vertical axis of the cylinder lies on a cylinder of radius Ut . The situation is illustrated in Fig. 5, which shows the positions of the gas cloud in three separate realizations, with C_1, C_2, C_3 denoting the positions of its axis in these realizations. In what follows, and in Fig. 5, it is assumed that $Ut > R$.

Now consider the ensemble of realizations in which x is measured relative to the fixed point O , the initial centre of mass of the gas cloud. Let P be a fixed point on the circle of radius Ut . The value of $p(\theta; x, t)$ at P depends on the probability distribution of the random direction of advection of the cloud. The simplest assumption is that this distribution is isotropic, that is, it is the same in all horizontal directions. In this case the proportion of realizations for which, at time t , P lies within the gas cloud is equal to $(\phi/2\pi)$, where ϕ is the angle (in radians) subtended at O by the gas cloud. By elementary trigonometry $\sin(\frac{1}{2}\phi) = (R/2Ut)$ so that, at P , and for this ensemble:

$$p(\theta; x, t) = \left[\frac{2}{\pi} \arcsin\left(\frac{R}{2Ut}\right) \right] \delta\left(\theta - \frac{Q}{\pi R^2 H}\right) + \left[1 - \frac{2}{\pi} \arcsin\left(\frac{R}{2Ut}\right) \right] \delta(\theta), \quad (14)$$

where δ is the ordinary delta function. The class of probability density func-

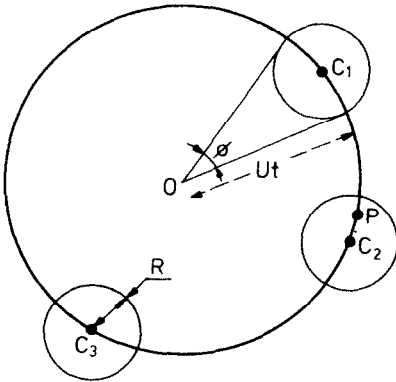


Fig. 5. Sketch illustrating discussion of use of box model in different ensembles.

tions like eqn. (14) is discussed by Chatwin and Sullivan [23]. From eqn. (8) it then follows that at P, and for this ensemble,

$$C(x, t) = \left(\frac{Q}{\pi R^2 H} \right) \left[\frac{2}{\pi} \arcsin \left(\frac{R}{2Ut} \right) \right]. \tag{15}$$

It can be seen immediately from eqn. (15) that $C(x, t)$, the ensemble mean concentration, is much less than $(Q/\pi R^2 H)$, the real concentration, whenever R is much less than Ut . According to most investigators using box models (van Ulden [22], Picknett [17] and Fay [8]), this condition will be satisfied since R becomes approximately proportional to $t^{1/2}$.

Now consider a second ensemble of realizations whose only difference from the first is that x is measured throughout each realization relative to the instantaneous centre of mass of the cloud. Then, each realization is identical and $p(\theta; x, t)$ is given by

$$p(\theta; x, t) = \begin{cases} \delta \left(\theta - \frac{Q}{\pi R^2 H} \right), & |x| \leq R; \\ \delta(\theta), & |x| > R. \end{cases} \tag{16}$$

Thus for this ensemble, again using eqn. (8),

$$C(x, t) = \begin{cases} \left(\frac{Q}{\pi R^2 H} \right), & |x| \leq R; \\ 0, & |x| > R; \end{cases} \tag{17}$$

in other words, for this ensemble, $C(x, t)$ is identical with the real concentration.

Apart from illustrating the point that the statistical properties of $\Gamma(x, t)$ depend on the choice of the ensemble to which they refer, the example

above illustrates one of the important differences between the analysis of turbulent diffusion phenomena in the framework of absolute diffusion (in which x is measured relative to a point fixed in space, as in the first ensemble above) and in the framework of relative diffusion (in which x is measured relative to a point moving with the dispersing cloud, as in the second ensemble above). When the framework of absolute diffusion is used, as in the first ensemble, the statistical properties of $\Gamma(x, t)$ depend significantly on those eddies of the turbulence that control the motion of the cloud as a whole ("meandering"). These eddies have large length and time scales and the experimental determination of the statistical properties of $\Gamma(x, t)$ requires that these scales are all fully incorporated in the data. Consequently, very many realizations of the dispersion will be needed to obtain reliable estimates of the statistical properties. Furthermore, the values of these properties will be very different from the corresponding real values and, in particular, the ensemble mean concentration will be much less than the real concentration as illustrated by eqn. (15). In contrast, when the framework of relative diffusion is used, as in the second ensemble, the choice of the origin of x ensures automatically that the meandering of the cloud as a whole does not affect the statistical properties. The only eddies of the turbulence that are relevant are those that influence the distortion of the gas cloud about its centre. The length and time scales of such eddies are comparable with those of the cloud itself, and are therefore normally much less than the scales of the meandering which are relevant in absolute diffusion. In principle, therefore, it is a much more practical proposition to determine the statistical properties of $\Gamma(x, t)$ in the framework of relative diffusion than in that of absolute diffusion. In many circumstances there is another advantage in using relative diffusion (Batchelor [15], Chatwin and Sullivan [3]). This is that the length scales of the controlling eddies in relative diffusion lie in the inertial subrange so that many powerful results follow from dimensional considerations. The eddies controlling the meandering do not lie in the inertial subrange. There are, however, two reasons why this advantage is unlikely to be of prime importance in heavy gas dispersion. These are the strong influence of buoyancy forces and the proximity of the ground.

Despite these arguments in favour of using relative, rather than absolute, diffusion as a framework, there is an obvious, and seemingly almost overwhelming, advantage in using absolute diffusion in practical hazard analysis. This advantage is that what matters is the location of the hazards relative to the container and other fixed buildings and topographical features. Thus the statistical properties of $\Gamma(x, t)$ are required for an ensemble of realizations in which the origin of x is fixed in space. Such information is given only when the framework of absolute diffusion is used.

The preceding discussion on different types of ensemble has, it is hoped, made clear the need for precise specification of the ensemble as an integral part of any hazard analysis. In particular, it should be realized that the ensemble for which practical predictions are required is not likely to be

that which is most convenient for conducting field trials. Thus, the hazard analysis for any particular container must consider all possible conditions of the ambient atmospheric turbulence, and the possibility of release occurring at any time of the day or night during any season of the year. On the other hand, field trials are likely to be conducted when the weather is fine and during the day time. Attention has therefore to be given to the extrapolation from the results of the field trials to the predictions for a real container.

One practical solution to this extrapolation problem seems possible when the terrain and the statistical properties of the ambient turbulence are the same in all horizontal directions (horizontal isotropy). In such a case, changes in the direction of the advection of the cloud as a whole ought not significantly to *increase* the probability that hazardous conditions exist at any point x at time after release t . Then each trial in a series is a typical realization of those in a hypothetical ensemble (to be used in the analysis of hazards) in which, in each realization of the ensemble, the cloud is advected in a direction which does not change during the realization, but which does change from realization to realization. The situation envisaged is very like that analyzed above and illustrated in Fig. 5, except that, instead of being constant, the speed of advection will be, in general, a random function of time. A hypothetical ensemble of this type can be constructed for each stability class and predictions for the ensemble of all possible releases can be obtained by making weighted combinations of the results from each separate ensemble, with the weights being based on the observed frequency of occurrence of each stability class. This procedure needs modification when horizontal isotropy is not satisfied because, for example, there are large buildings on one side of the container; also it does not take account of changes in the stability class during dispersion.

The effects of finite resolution on experimental measurements of concentration

It has already been explained in the Introduction to this paper that it is normally impossible to measure $\Gamma(x, t)$ in real experiments. Because of the finite size of concentration sensors, and the finite time taken to make measurements, every experimental value of concentration is inevitably an average of $\Gamma(x, t)$ over a certain region D of space-time. Because of the construction and mode of functioning of the sensor this average may be weighted if, for example, different parts of the sensor respond differently than others to the gas-air mixture. Thus the value of $\Gamma(x, t)$ that is recorded, denoted by $\Gamma_m(x, t)$, will satisfy an equation of the form

$$\Gamma_m(x, t) = \frac{\int_D w(x', t') \Gamma(x', t') dV(x') dt'}{\int_D w(x', t') dV(x') dt'} \quad (18)$$

where both the weight function w and the region D are characteristics of the sensor, to be determined by calibration. The integration over D involves both

an integration over a volume Δ containing \mathbf{x} , and an integration over a time interval τ containing t .

Significant differences between $\Gamma(\mathbf{x}, t)$ and $\Gamma_m(\mathbf{x}, t)$ are likely to arise whenever the size of the volume Δ and the time interval τ (which together form the region D) are not small compared with all scales characteristic of the structure and variation of the actual distribution of concentration $\Gamma(\mathbf{x}, t)$. The smallest length scale will normally be of the order of the conduction cut-off length λ_c (whose definition, and physical significance, is discussed in, for example, Chatwin and Sullivan [3]), and the smallest time scale of the order of (λ_c/U) , where U is a typical advection velocity at the location of the sensor. Differences between $\Gamma(\mathbf{x}, t)$ and $\Gamma_m(\mathbf{x}, t)$ could therefore be expected to be small whenever

$$\Delta^{1/3} \ll \lambda_c \quad \text{and} \quad \tau \ll (\lambda_c/U). \quad (19)$$

Taking typical values in field trials of λ_c and U as 10^{-3} m and 1 m sec $^{-1}$, respectively, in eqn. (19), gives $\Delta^{1/3} \ll 10^{-3}$ m and $\tau \ll 10^{-3}$ sec, conditions which seem almost impossible to meet in practice. In the trials reported by Picknett [17], for example, the gas sensors used had $\Delta^{1/3}$ of the order of 10^{-1} m and τ of the order of 5×10^{-2} sec.

The primary consequence of the seemingly inevitable differences between $\Gamma(\mathbf{x}, t)$ and $\Gamma_m(\mathbf{x}, t)$ in field trials is that the distribution of $\Gamma_m(\mathbf{x}, t)$ will not represent correctly the fine-scale structure actually present in the distribution of $\Gamma(\mathbf{x}, t)$. Consider a simple example with a particular sensor for which Δ is a cube of side 10^{-1} m, τ is negligibly small and w , the weight function in eqn. (18), is 1 everywhere. Let 90% of Δ be pure air and the remaining 10% be a gas-air mixture mainly occupying volume elements of minimum dimension of order λ_c . Suppose that the concentration of gas $\Gamma(\mathbf{x}, t)$ within the gas-air mixture is 10%. The measured concentration of gas $\Gamma_m(\mathbf{x}, t)$ will however be 1%, giving no indication that flammable conditions exist within Δ (assuming that the gas is methane). While this example is over-simple, because it ignores the smoothing effects on the distribution of actual concentration $\Gamma(\mathbf{x}, t)$ of both non-uniform initial mixing and of molecular diffusion, it does illustrate that care is needed before assessing hazards in terms of $\Gamma_m(\mathbf{x}, t)$.

However it should also be particularly stressed now that $\Gamma_m(\mathbf{x}, t)$, like $\Gamma(\mathbf{x}, t)$, is a random variable to which the statistical description developed earlier in this paper can be directly applied. A probability density function $p_m(\theta; \mathbf{x}, t)$ can be defined by eqn. (5) with $\Gamma(\mathbf{x}, t)$ replaced by $\Gamma_m(\mathbf{x}, t)$. Likewise the ensemble mean measured concentration $C_m(\mathbf{x}, t)$ and the ensemble mean square measured fluctuation $c_m^2(\mathbf{x}, t)$ can be defined by equations analogous to (8) and (9) respectively. In general it is clear that, unless eqn. (19) is satisfied, statistical properties of $\Gamma(\mathbf{x}, t)$ depending significantly on its fine-scale structure will not be closely estimated by the corresponding properties of $\Gamma_m(\mathbf{x}, t)$.

Using $p_m(\theta; x, t)$, a quantity $P_m(x, t)$ can be defined by an equation like (11), and hence, with one important proviso, the statistical description of hazards developed above can be used with $p_m(\theta; x, t)$ instead of $p(\theta; x, t)$. The important proviso is that it must be possible to express satisfactorily the condition that a hazard is present at position x and time t in terms of the measured concentration $\Gamma_m(x, t)$, rather than the actual concentration $\Gamma(x, t)$. Thus the simple example above and the experimental evidence of Birch, Brown and Dodson [16], illustrated in Figs. 2, 3 and 4, suggest that, because of the fine-scale heterogeneity in the structure of the actual concentration $\Gamma(x, t)$, the relation

$$0.05 < \Gamma_m(x, t) < 0.15 \quad (20)$$

may not be a satisfactory criterion for assessing whether flammable conditions exist at position x and time t in a cloud of methane dispersing in air. Investigation is needed for each hazard (and for each set of sensor characteristics) to determine whether a simple criterion like eqn. (20) can be established.

Provided this proviso can be met, it becomes clear that the apparent disadvantages of the differences between $\Gamma(x, t)$ and $\Gamma_m(x, t)$ need not be a major obstacle, though it is clearly important that, in any report of experimental data, there should be clear and full statements of the sensor characteristics.

The experimental estimation of concentration fluctuations

While the main theme of this paper has been that research, both experimental and theoretical, should aim at estimating $p(\theta; x, t)$, it is clear that this is not easy. A less ambitious aim (but one which should be regarded as second-best) is to estimate the distribution of $\overline{c^2}(x, t)$, the mean square fluctuation. This provides the simplest measure of the variability that can be expected between two realizations of the dispersion, and it is clear that some such measure is essential in any proper hazard analysis since this cannot be based on the mean concentration $C(x, t)$ alone.

The statements made above can be quantified when the form of $p(\theta; x, t)$ is known. For example, evidence like that shown in Fig. 1(a) suggests that $p(\theta; x, t)$ may be well approximated by a Gaussian function of θ near the centre of the cloud. Then the probability that $\Gamma(x, t)$, the concentration in any one trial, differs from $C(x, t)$ by more than $\sqrt{\overline{c^2}(x, t)}$ is $2\{1 - \Phi(1)\} \approx 0.317$, where Φ is the cumulative distribution function for the Gaussian distribution. An inverse type of calculation can also be made, namely the determination of concentration confidence limits between which the actual concentration has a specified probability of lying. Thus, for example, since $\Phi(1.65) \approx 0.95$, there is a 90% chance that $\Gamma(x, t)$ lies between the confidence limits $C(x, t) \pm 1.65\sqrt{\overline{c^2}(x, t)}$. Both of these types of calculation can be made when $p(\theta; x, t)$ has a form other than Gaussian, provided $C(x, t)$ and $\overline{c^2}(x, t)$ can be estimated.

It was noted earlier that there are many models which aim at predicting the ensemble mean concentration $C(x, t)$. Measurements of $\Gamma(x, t)$ then enable estimates of $c(x, t)$ to be made, using eqn. (2). It is known (Chatwin and Sullivan [24]) that there are several general results that $c(x, t)$ must satisfy such as

$$\int c(x, t) dV(x) = \int c(x, t) c(x + y, t) dV(x) = 0, \quad (21)$$

where the integrals are over the whole region occupied by the dispersing gas. If the experimental estimates of $c(x, t)$ do not satisfy relations like those in eqn. (21), adjustments can be made to the theoretical estimates of the ensemble mean concentration. There are other relations, given in Chatwin and Sullivan [3, 4], which $\bar{c}^2(x, t)$ must satisfy, and these can also be used as checks of the experimental estimates.

Concluding remarks

For reasons of space, this paper contains no discussion of two important topics which are considered elsewhere [25]. These are the influences on $p(\theta; x, t)$ of the basic physical processes governing dispersion, such as advection, molecular diffusion and the effect of buoyancy forces, and a discussion of simple forms of $p(\theta; x, t)$ that might be practically adequate for hazard analysis. It is hoped however that the exclusion of these topics will have helped to stress the merits and practical usefulness of the statistical approach which this paper has tried to summarize.

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References

- 1 G.T. Csanady, *Turbulent Diffusion in the Environment*, Reidel, Dordrecht, 1973.
- 2 J.E. Fackrell and A.G. Robins, Concentration fields associated with emissions from point sources in turbulent boundary layers, Part II — the effects of source size, C.E.G.B. Report MM/MECH/TF 264, Marchwood Engineering Labs., Southampton, 1980.
- 3 P.C. Chatwin and P.J. Sullivan, The relative diffusion of a cloud of passive contaminant in incompressible turbulent flow, *J. Fluid Mech.*, 91 (1979) 337.
- 4 P.C. Chatwin and P.J. Sullivan, Measurements of concentration fluctuations in relative turbulent diffusion, *J. Fluid Mech.*, 94 (1979) 83.
- 5 J.A. Havens, Predictability of LNG vapor dispersion from catastrophic spills onto water: an assessment, U.S. Coast Guard Report No. CG-M-09-77, Washington, DC 1977.
- 6 J.A. Havens, A description and assessment of the SIGMET LNG vapor dispersion model, U.S. Coast Guard Report No. CG-M-3-79, Washington, DC, 1979.

- 7 J. McQuaid, Dispersion of heavier-than-air gases in the atmosphere, Health and Safety Executive Technical Paper 8, HSE, Sheffield, 1979.
- 8 J.A. Fay, Gravitational spread and dilution of heavy vapor clouds, Proc. 2nd International Symp. on Stratified Flows, Trondheim, Norway, 1980, p. 471.
- 9 A.S. Monin and A.M. Yaglom, Statistical Fluid Mechanics, Vol. 1, MIT Press, Cambridge, MA, 1971.
- 10 A.S. Monin and A.M. Yaglom, Statistical Fluid Mechanics, Vol. 2, MIT Press, Cambridge, MA, 1975.
- 11 T.S. Lundgren, Distribution functions in the statistical theory of turbulence, Phys. Fluids, 10 (1967) 969.
- 12 A.S. Monin, Equations for finite dimensional probability distributions of a field of turbulence, Dokl. Akad. Nauk SSSR, 177 (1967) 1036.
- 13 G.I. Taylor, Diffusion by continuous movements, Proc. Lond. Math. Soc., (2) 20 (1921) 196.
- 14 L.F. Richardson, Atmospheric diffusion shown on a distance-neighbour graph, Phil. Trans. Roy. Soc. A, 110 (1926) 709.
- 15 G.K. Batchelor, Diffusion in a field of homogeneous turbulence, II, The relative motion of particles, Proc. Camb. Phil. Soc., 48 (1952) 345.
- 16 A.D. Birch, D.R. Brown and M.G. Dodson, Ignition probabilities in turbulent mixing flows, Report No. MRS E 374, Midlands Research Station, British Gas, Solihull, 1980.
- 17 R.G. Picknett, Field experiments on the behaviour of dense clouds, Report No. Ptn. IL 1154/78/1, Chemical Defence Establishment, Porton Down, Wiltshire, 1978; see also Dispersion of dense gas puffs released in the atmosphere at ground level, Atm. Env., 15 (1981) 509.
- 18 C. Dopazo and E.E. O'Brien, Statistical treatment of non-isothermal chemical reactions in turbulence, Comb. Sci. & Tech., 13 (1976) 99.
- 19 J. Janicka, W. Kolbe and W. Kollmann, Closure of the transport equation for the probability density function of turbulent scalar fields, J. Non-Equil. Thermodyn., 4 (1979) 47.
- 20 S.B. Pope, The statistical theory of turbulent flames, Phil. Trans. Roy. Soc. A, 291 (1979) 529.
- 21 A.D. Birch, D.R. Brown, M.G. Dodson and J.R. Thomas, The turbulent concentration field of a methane jet, J. Fluid Mech., 88 (1978) 431.
- 22 A.P. van Ulden, On the spreading of a heavy gas released near the ground, In: C.H. Buschmann (Ed.), Proc. 1st International Symposium on Loss Prevention and Safety Promotion in the Process Industries, Elsevier, Amsterdam, 1974, p. 221.
- 23 P.C. Chatwin and P.J. Sullivan, The structure of the probability density function of concentration in turbulent diffusion, Proc. 13th A.I.A.A. Fluid and Plasma Dynamics Conference, Snowmass, CO, 1980.
- 24 P.C. Chatwin and P.J. Sullivan, Some turbulent diffusion invariants, J. Fluid Mech., 97 (1980) 405.
- 25 P.C. Chatwin, The influence of basic physical processes on the statistical properties of dispersing heavy gas clouds, Proc. 7th Biennial Symposium on Turbulence, Rolla, MO, 1981.