INVITED

Modelling the Hazards Posed by the Sudden Release of a Quantity of Contaminant

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Abstract The consequence of an accidental release of toxic, flammable or noxious material into an environmental flow needs to be assessed in terms of the territory that is subjected to contaminant at unacceptable concentration levels. Environmental flows are turbulent and generally unsteady and inhomogeneous such that the contaminant concentration values in question are nonstationary, inhomogeneous random variables. Thus the practically important, basic, problem of describing the evolution of the concentration field in a contaminant cloud presents some serious theoretical and experimental challenges. A new approach is developed to describe the diffusion of a contaminant cloud in terms of its location, size and state. The state of the cloud will characterize the dilution of the contaminant concentration values within the cloud. A new measure - the 'expected mass fraction' function - is introduced to describe the state of cloud dilution. The advantages of this approach are discussed in terms of the experimental difficulties associated with taking averages. The theoretical advantages that follow from this new approach are illustrated in terms of recent, simple models of the evolution of the moments of the one-point probability density function of concentration.

INTRODUCTION

The objective in this paper is to address issues of contaminant diffusion that are peculiar to an environmental flow. The focus will be on the sudden release of a contaminant fluid into a miscible host fluid. This is justified in that most accidental releases occur in this way. The problem then is to characterize the reduction of contaminant concentration values $\Gamma(\mathbf{x},t)$ found at the position located by vector $\mathbf{x}$ at time t in the contaminant cloud formed by the conserved, initial release mass Q.

In a turbulent velocity field the concentration Γ is an intrinsically nonstationary and inhomogeneous random variable and in addition, environmental flows are generally unsteady, three dimensional, shear flows. Although turbulent convective motions rapidly transport contaminant over large distances the actual reduction of contaminant concentration takes place more slowly, on a very much smaller spatial scale, through molecular diffusivity κ. The conventional representation of concentration reduction is with the probability density function $p(\theta;\mathbf{x},t)$ where,

$$p(\theta;\mathbf{x},t)d\theta = \text{prob}\{\theta \le \Gamma(\mathbf{x},t) < \theta + d\theta\}. \qquad (1)$$

In the case of contaminant clouds averages are formed from many repetitions of mass release. The reasonably well controlled laboratory experiment of Hall et al (1991), where up to 100 repetitions were used, suggest that measurement of the pdf for a contaminant cloud would be impractical. Most of this paper will be devoted to a description of a new measure of contaminant concentration reduction in clouds - the expected mass fraction function (c.f. Chatwin and Sullivan 1990a, Sullivan and Ye 1993).

One issue that immediately arises in unsteady environmental flows is the dependence of the measure on the different time and length scales of turbulent motion. For example the cloud size as observed in an inertial or fixed reference frame in a homogeneous turbulent field is determined by the largest scales of turbulent motion (c.f. Batchelor 1949). It is conceivable that the time change of speed and or direction of the flow field could be comparable with the largest time scales of the turbulence. It is therefore important to attempt to isolate phenomena in terms of the spectral scales on which they depend. In general, there is likely to be a limited amount of detailed information available in terms of accidental release conditions or flow conditions such that a consistently simple and robust representation is developed in the following sections.

DECOMPOSITION

It is useful to view the evolving contaminant cloud in terms of its location, size, and, state of concentration values. The location is given by the center-of-mass position vector $\mathbf{X}_c$ and the size by a vector $\mathbf{Y}$ representing the spatial variance in center-of-mass coordinates. That is for one component,

$$y_i = Q^{-1}\int_{a.s.}(x_{ci}-x_i)^2\Gamma(\mathbf{x},t)dv \qquad (2)$$

where a.s. designates an integral over all space. The 'state' of the contaminant concentration field will be given, in each realization, by the mass fraction function $\hat{p}_i(\theta;t)$ which will be developed in the next section. Thus the joint probability of $\mathbf{X}_c$,

$\mathbf{Y}$ and $\hat{p}_i(\theta;t)$ will charactarize the contaminant field. The location and size of the cloud are virtually independent of molecular diffusivity κ and hence do not address the issue of concentration reduction.

Generally, the location is determined by the largest scales of turbulent motion while the size is determined by scales comparable with the cloud size. For large Reynolds number environmental flows, away from boundaries, the scales that govern the location and size are statistically decoupled and indeed the average spatial variance growth rate for the isotropic inertial subrange is simply given by (c.f. Batchelor 1951)

$$y^2 = \gamma \epsilon t^3, \tag{3}$$

where γ is a universal order one constant and ε is the rate of turbulent energy dissipation per unit mass. Turbulent convective motions pull out the contaminant mass into thin sheets and strands until the thinning is balanced by molecular diffusion at the conduction cut-off length $\lambda = (\kappa^2\nu/\epsilon)^{1/4}$, where ν is the kinematic viscosity. It should be noted that λ is 10^{-3} – 10^{-5}m in most flows and, although turbulent convective motions will spread a cloud over kilometers in environmental flows, the mixing due to κ occurs over the very small λ dimension. Thus one anticipates a further statistical decoupling of the turbulent scales that contribute to concentration reduction from those that determine location and size. When this is true the charactarization will consist of considering separately the probable location, size and state of the cloud. Practically, this would amount to considering the average value and the variability (which in general will decrease in the order) of $\mathbf{X}_c$, $\mathbf{Y}$ and $\hat{p}_i(\theta;t)$. The anticipated, relatively low, variability of the mass fraction function $\hat{p}_i(\theta;t)$ was one of the principle motivations for its development which now follows.

THE EXPECTED MASS FRACTION

The expected mass fraction fuction is conceptually straightforward. At a given time following the release of mass Q, all of the continuum-scale volume elements containing concentration values in the range $\theta \le \Gamma < \theta + d\theta$ are summed and the total mass in that concentration range compiled. This process is repeated and a numerical average taken over many releases and the result normalized by the conserved release mass Q. Thus, on average, the distribution of release mass over the concentration range $[0,\theta_0]$, where θ_0 is the highest value of concentration at release, is portrayed as a function of time following release. The reduction in contaminant concentration values is apparent through the transfer of mass to lower values of concentration as time increases.

The expected mass fraction function is defined in terms of the pdf as,

$$\tilde{p}(\theta;t) = Q^{-1}\int_{a.s.} \theta p(\theta;\mathbf{x},t)dv\,, \tag{4}$$

such that,

$$\int_{\theta_a}^{\theta_b} \tilde{p}(\theta;t)d\theta\,, \tag{5}$$

is, on average, the fraction of release mass found on the concentration interval between θ_a and θ_b at time t and,

$$\int_0^{\theta_0} \tilde{p}(\theta;t)d\theta = 1\,. \tag{6}$$

Intuitively, the variability of the mass fraction function $\hat{p}_i(\theta;t)$ about the expected mass fraction function $\tilde{p}(\theta;t)$ is small since (a) there is no spatial reference involved and (b) an integral over the concentration values in the entire cloud is used in each realization. Unfortunately, there is very little data on the important cloud problem and none suitable for the exploration of $\hat{p}(\theta;t)$. An experimental design for a useful experiment for this purpose is found in Ye (1995).

A fixed-point version of (6) is developed in Heagy and Sullivan (1995a). There the values of concentration that are observed as a cloud passes a fixed sampling position, say $\mathbf{x}_0$, are used to compile,

$$\tilde{p}(\theta;\mathbf{x}_0) = \bar{d}^{-1}\int_0^{\infty} \theta p(\theta;\mathbf{x}_0,t)dt, \tag{7}$$

where,

$$d(\mathbf{x}_0) = \int_0^{\infty} \Gamma(\mathbf{x}_0,t)dt, \tag{8}$$

and an overbar is used to denote an ensemble average. It was shown that the variability about $\tilde{p}(\theta;t)$, from realization to realization, was comparable with that of the low-information statistic, dosage, $\bar{d}(\mathbf{x})$. Of course the variability about $\tilde{p}$ is far larger than that anticipated for $\hat{p}$, however, small enough that the measured values from Hall et al (1991) were reasonably convergent.

The representation of concentration values or 'state' of the cloud using $\tilde{p}(\theta;t)$ provides a measure with a straightforward interpretation which is not subjective (c.f. discussion of intermittency by comparison in Chatwin and Sullivan 1989). The expected mass fraction function provides a compromise

containing less information than $p(\theta;x,t)$ but which does represent the reduction in concentration values through κ which, for example, $\overline{\Gamma}(x,t)$ does not. In addition to the desirable features of marshalling relevent information into a readily interpretable format and of rapid statistical convergence, $\hat{p}(\theta;t)$ offers some theoretical advantages which will now be explored.

MOMENTS

A useful way of approximating a pdf (or in this case $\hat{p}(\theta;t)$) is to use measured or calculated lower moments with an inversion proceedure, such as a maximum entropy method (Derksen and Sullivan 1990 and Derksen, Sullivan and Yip 1994). The integral moments of $\hat{p}(\theta;t)$,

$$\hat{M}_n(t) = \int_0^\infty \theta^n \hat{p}(\theta;t)d\theta, \tag{9}$$

are simply related to the moments of $p(\theta;x,t)$,

$$m_n(x,t) = \int_0^\infty \theta^n p(\theta;x,t)d\theta \tag{10}$$

as,

$$\hat{M}_n(t) = Q^{-1}\int_{a.s.} m_{n,1}(x,t)dv. \tag{11}$$

A similar simple expression is obtained for the fixed-point definition (7),

$$\tilde{M}_n(x_0) = \bar{d}^{-1}\int_0^\infty m_{n+1}(x_0,t)dt, \tag{12}$$

and where $\tilde{M}(x_0)$ can be simply compiled experimentally from,

$$\tilde{M}_n(x_0) = \bar{d}^{-1}\int_0^{\overline{\infty}} \Gamma^n(x_0,t)dt. \tag{13}$$

Further, an exact expression for $\hat{M}_n(t)$,

$$\hat{M}_n(t) = -n(n+1)\kappa\int_0^t f(t')dt' + \hat{M}_n(0), \tag{14}$$

where,

$$f(t) = Q^{-1}\int_{a.s.} \overline{\Gamma^{n-1}(x,t)(\nabla\Gamma(x,t))^2}dv, \tag{15}$$

is given in Chatwin and Sullivan (1990a).

The advantage of (15) is that one can affect closure on the integrand where integration is over all of space. This is in contrast to the normal procedure of replacing terms in the non-linear partial differential equations that govern the process of contaminant concentration reduction (c.f. discussion in Mole, Chatwin and Sullivan 1993, Chatwin and Sullivan 1994).

An expression for the distributed central moments,

$$\mu_n(x,t) = \int_0^\infty (\theta - \overline{\Gamma(x,t)})^n p(\theta;x,t)d\theta, \tag{16}$$

was given in Chatwin and Sullivan (1990b) as,

$$\begin{aligned}\mu_n(x,t) = \beta_n^n(t)\{&m_1(x,t)(\alpha_n(t)m_1(c,t)-m_1(x,t))^n \\ &+ (-1)^n(\alpha_n(t)m_1(c,t)-m_1(x,t))m_1^n(x,t)\},\end{aligned} \tag{17}$$

where c is the position where the mean concentration takes a maximum value. The formulation of moments in (17) has received a respectable amount of direct experimental validation in a wide variety of steady turbulent flows and notably in Sawford and Sullivan (1995). In general the dependence of β_n and α_n on n was observed to be slight. By assuming α_n and β_n to be independent of n, Mole and Clarke (1995) developed the parameter free relationship between kurtosis K and skewness S from (17) to be

$$K = S^2+1, \tag{18}$$

which is a lower bound for any pdf and corresponds to a two state process. Ye (1995) has shown, using Mellin transforms, that the pdf corresponding to (17) with $\beta_n = \beta$ and $\alpha_n = \alpha$ is,

$$\left.\begin{aligned} &p(\theta;x,t) = p_1\delta(\theta-\theta_1) + (1-p_1)\delta(\theta-\theta_2), \\ &\text{where,} \\ &\theta_1 = \beta(t)\alpha(t)m_1(c,t) + (1-\beta(t))m_1(x,t), \\ &\theta_2 = (1-\beta(t))m_1(x,t), \\ &\text{and} \\ &p_1 = \frac{m_1(x,t)}{\alpha(t)m_1(c,t)}. \end{aligned}\right\} \tag{19}$$

The data from a wide variety of laboratory and field experiments, including flows with obstacles in the flow field, have been shown to collapse onto a curve that is close to, but with kurtosis values slightly in excess of, that given in (18). In particular, albeit indirectly, the collapse of cloud data (Heagy and Sullivan 1995b) suggests that (17) is also appropriate there as well.

Ye (1995) has provided physical arguments for the inclusion of a convolution function with (19). To a very good approximation, over the bulk of the experimental flows considered (a point and a line source in grid turbulence and the cross flow from a buoyant, round jet protruding into a wall boundary layer), the convolution function was found to be Gaussian leading to the bi-Gaussian pdf,

$$p(\theta) = \frac{1}{\sigma\sqrt{2\pi}}\left[p_1 \exp\left\{\frac{-(\theta-\theta_1)^2}{2\sigma^2}\right\} + (1-p_1)\exp\left\{\frac{-(\theta-\theta_2)^2}{2\sigma^2}\right\}\right], \tag{20}$$

where $\sigma = am_1(c,t)$ with $.08 < a < .13$. Equation (20) provides an improved fit, over (18), to experimental data (Sullivan and Ye 1995) and accounts for the observed, slight, dependence of α_n and β_n on n.

By considering the simple and plausible closure approximation,

$$(\nabla\Gamma)^2 = A\left(\frac{\Gamma-\Gamma_t}{\lambda}\right)^2, \tag{21}$$

where $\Gamma_t \to 0$ as $t \to 0$, $\Gamma_t \to \overline{\Gamma}$ as $t \to \infty$, and A a closure constant, Labropulu and Sullivan (1995) have derived the rather general expression for $\alpha_2(t)$ and $\beta_2(t)$ to be,

$$\left.\begin{aligned} \hat{\alpha} &= \frac{-e^{\tau}}{Qm_1'(0,\tau)}\int_0^{\tau} e^{-\tau'}\frac{d}{d\tau'}\int_{a.s.} m_1^2(x,\tau')dv\,d\tau' \\ &\text{where,} \\ \hat{\alpha} &= \alpha_2 - \frac{1}{Qm_1(0,\tau)}\int_{a.s.}\mu_2 dv, \\ \beta_2^2 &= \frac{\theta_0}{m_1(0,\tau)m_1'(0,\tau)}\frac{dm_1(0,\tau)}{d\tau}; \quad \tau = \frac{2\kappa A}{\lambda^2}t. \end{aligned}\right\} \tag{22}$$

Equation (22) with modifications to apply to steady homogeneous and shear flows has been shown to (at least qualitatively) represent the limited amount of experimental data available. Mole and Clarke (1995) provide experimental field data that support the use of (21). They have used a modified form of (21) with (14), assuming α_n and β_n to be independent of n for n=2 and n=3 to obtain numerical solutions for the non-linear ordinary differential equations so obtained. The $\alpha(t)$ and $\beta(t)$ values they derived appear to be qualitatively similar to those given in (22).

One can use the $\alpha_2(t)$ and $\beta_2(t)$ values predicted by (22) to determine θ_1, θ_2 and p_1 from (19) and hence, assuming a to be constant, $p(\theta;x,t)$ from (20). This is then used in (4) and (7) to obtain $\tilde{p}(\theta;t)$ and $\tilde{p}(\theta;x_0)$. An illustative example calculation in the neutral atmospheric boundary layer using Lagrangian similarity analysis is shown for $\tilde{p}(\theta;t)$ in Sullivan and Ye (1995) and for $\tilde{p}(\theta;x_0)$ in Heagy and Sullivan (1995).

It is worth noting that the fixed point prediction of $\mu_2(x_0,t)$ using (22) and (17) and $\tilde{p}(\theta;x_0)$ could be measured in a suitably designed wind tunnel experiment. A general comparison with the Hall et al (1991) data looks reasonable and in particular the shapes and relative magnitudes of $\overline{\Gamma}(x_0,t)$ and $\mu_2(x_0,t)$, however, the release conditions prohibit detailed comparison. There is a critical need for reliable experimental data. Some reasons why reliable data aquisition is problematic are explored in the following section.

CONCENTRATION MEASUREMENTS

Different applications emphasize different segments of the pdf of concentration. Odour detection involves parts per billion of contaminant and emphasizes the extreme values found in the high concentration pdf tails, whereas combustion focuses on generally lower values within the flammability limits of concentration. Toxicity may in general depend nonlinearly on contaminant concentrations. Some recent work on steady plumes show promise in representing the tails of the pdf with a Pareto distribution (Lewis and Chatwin 1995a and Wright, Anderson and Mole 1995) and in dealing with the "thresholding" problem (distinguishing low level concentration values from instrument noise) using a maximum entropy procedure (Lewis and Chatwin 1995b, Robinson, Lewis and Chatwin 1995).

The epoch over which samples are taken in environmental flows is important because of the general unsteadiness of these flows and the filtering effect of excluding relevent information (c.f. recent work in Echman 1994). Fixed-point measurements are especially vulnerable to this problem.

In addition to the above difficulties, there is an instrument smoothing problem in both field and laboratory measurements that arises due to the contaminant being confined to sheets and strands of the very small conduction cut off dimension λ. It is to be noted that the mean concentration $\overline{\Gamma}(x,t)$ is not sensitive to either spatial and temporal averaging i.e.,

$$\bar{I} = \frac{1}{VT}\overline{\int_V \int_T \Gamma(x,t)dv\,dt} = \bar{\Gamma}(x,t), \qquad (23)$$

or the effects of molecular diffusion κ, however, all higher moments are very sensitive to (and are reduced by) instrument smoothing.

The effects of time averaging have been clearly shown to be important by Mylne and Mason (1991) using deconvolution and in the analysis of Derksen and Sullivan (1987) and Sullivan (1984). The importance of spatial averaging was discussed in Chatwin and Sullivan (1993) where fine scale measurements of Sakai et al of the second central moments of concentration on the center line of an experimental jet virtually doubled as their sample volume was decreased from $(0.54mm)^3$ to $(0.10mm)^3$.

It appears unlikely that a robust probe for use in field measurement which is not subject to instrument smoothing will be developed in the near future. To overcome this difficulty an extrapolative technique (manuscript in preparation with T. Shopflocher) is proposed to obtain better estimates from spatially averaged concentration measurements. A statistically isotropic concentration field is assumed over a small sample probe volume and the ratio of continuum-scale-resolved central moment to the measured value developed as a power series in the sample volume size. The first few coefficients of that series are then determined from measured moments using different sample volumes. This framework, when applied to the Sakai et al data (with only 4 different sample volumes used in the measurements) provides reasonable results. One can use corrections to the first four measured moments and, through the inversion technique mentioned in the Introduction, derive a corrected pdf estimate.

Given the complex nature of turbulent diffusion, one relies very heavily on measured information. There is sufficient question surrounding the interpretation of measured concentration moments (c.f. Corriveau and Baines 1993 and Dahm et al 1991 for direct evidence of the texture of the contaminant field) with respect to spatial averaging to warrant a full scale experimental investigation.

CONCLUDING REMARKS

The key issue in modelling the hazard or risk associated with the sudden release of contaminant is to establish the likelihood of encounter of problematical concentrations at a given time and spatial location. The decomposition of the problem into one of considering the probable location, size and state of the cloud provides a flexible approach that accomodates unsteady flows (for example on-shore, off-shore island breezes) and situations where 'cloud meander' may or may not be important or in circumstances when one chooses not to attempt distinction between large scale turbulent motion and dynamic meteorological change. The decomposition is modest in the information it attempts to supply which is consistent with what is likely to be known about flow and release conditions or what information can actually be measured for essential validation. In particular it should be noted that the definition of the expected mass fraction function requires only the release mass to be conserved. This restriction can be removed by allowing Q to be a random function of time t and normalizing with $\overline{Q}$ in the definition. In order to make further progress, information from well designed and targeted laboratory and (subsequent) field experiments is required. Similar remarks apply to the physiological impact of different contaminants with respect to the fluctuations in concentration levels.

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