The need for a new measure of contaminant cloud concentration reduction(*)(**)

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Summary. — In this paper it will be demonstrated that conventional measures used to characterize contaminant concentration reduction are impractical and inadequate to describe the important case of a contaminant cloud. A new measure, the expected-mass-fraction function, is developed and shown to have desirable experimental and theoretical features. Some contaminant plume data is used to illustrate the application of this new measure.

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1. – Introduction

The contaminant cloud problem is important since many industrial mishaps including the rupture of storage containers, either fixed or in use in transportation, result in the sudden release of contaminant fluid. The problem considered here is to describe the reduction of contaminant concentration values when a quantity of contaminant is released in a miscible, turbulent, host fluid. Of particular concern will be the release of contaminant into an environmental flow such as the Earth's atmosphere, oceans, lakes and rivers.

The flow field is turbulent and the concentration $\Gamma(\mathbf{x}, t)$, in mass per unit volume at the position located by the vector $\mathbf{x}$ at time t, is a random variable. The root-mean-square value of concentration is, at least, comparable with the mean value in turbulent flows. The equations that determine the velocity and concentration fields are not "closed". There is no *a priori* justification for any closure hypothesis so that any semiempirical approximation or simulation must be thoroughly validated by experiment. In the case of steady

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laboratory flows and contaminant release ensemble averages are found from measured, stationary, time-series. By contrast, with a contaminant cloud, averages must be formed from the records obtained from the repeated release of contaminant with the same initial configuration. Even in the most idealized, steady, isotropic, homogeneous, turbulent field the concentration in a cloud is an inhomogeneous and nonstationary random variable. Environmental flows, in addition, are generally time and space dependent. Because we cannot compile ensemble averages in an environmental flow, not all statistics can be measured (or even reasonably approximated). For example, the epoch over which measurements are gathered must be adequate to compile the required statistic yet small with respect to significant changes in the turbulent field.

The only agency to reduce concentration values is molecular diffusivity κ. Turbulent convective motions pull out the contaminant into thin sheets and strands until the thinning is balanced by thickening due to molecular diffusion at the conduction cut-off length $\lambda = (\nu\kappa^2/\epsilon)^{\frac{1}{4}}$, where ν is the kinematic viscosity and ϵ is the rate of turbulent energy dissipation per unit mass. λ is of the order 10^{-3}–10^{-5} m in most flows. That is a contaminant cloud may be spread out over a length scale of the order of kilometres in an environmental flow but mixing between host and contaminant fluid takes place on the small λ length scale due to κ. Recent direct observational evidence of this sheet and strand texture of the contaminant field is given in Corriveau and Baines [1] and Dahm *et al.* [2]. The fact that contaminant is confined to these very thin sheets and strands gives rise to serious experimental temporal and spatial resolution problems and evidence of this is presented in Chatwin and Sullivan [3].

The natural way to charactarize contaminant concentration is with the one-point probability density function, PDF,

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

which has moments defined as

$$(2) \qquad\qquad m_n(\mathbf{x}, t) = \overline{\Gamma(\mathbf{x}, t)^n} = \int_0^\infty \theta^n p(\theta; \mathbf{x}, t)\, \mathrm{d}\theta,$$

where an overbar is used to denote an ensemble average. It is clear that the higher moments (large n in (2)) more heavily weight the relatively rare events that are described by the "tails" of the PDF and that a longer time series record in a steady experiment, or a larger number of realizations for clouds, is required for adequate approximations of their ensemble average values. It is worth noting that the lowest-order moment—the 'mean' $m_1(\mathbf{x}, t)$—is reasonably insensitive to space and time averaging (whereas all, non-zero, higher moments are significantly reduced by instrument smoothing). The mean concentration $m_1(\mathbf{x}, t)$ is also virtually insensitive to molecular diffusivity (whereas all higher moments are reduced by κ). Although it is the most reliably measured and easily predicted statistic, the mean conveys no information on mixing between host and contaminant fluid. Practical advantages were demonstrated, in Derksen and Sullivan [4] and Derksen, Sullivan and Yip [5], in using the lower-order measured moments (say the first four) and an inversion procedure, such as a maximum entropy formalism, to obtain an approximation of the PDF. This approach will be used later in this paper.

In a well-controlled laboratory experiment Hall *et al.* [6] made up to 100 repeat releases of dense gas clouds into the logarithmic boundary layer over their wind-tunnel floor. It was found that only the mean concentration $m_1(\mathbf{x}_0, t)$ (where $\mathbf{x}_0$ is a fixed sampling position) was reasonably convergent. It would be impractical, even in a steady laboratory flow, to measure the PDF for clouds.

One further point must be made on the distinctiveness of contaminant concentration reduction in clouds. Contaminant found in the sheet-strand structure described above is spread out over a region of space by large-scale turbulent convective motions. Concentration values are reduced through molecular transfer from high sheet and strand values to lower ambient concentrations found between the sheets and strands. The difference in the 'space-filling' nature of diffusing contaminant in a three-dimensional cloud from the case of a steady axisymmetric point or line source will very likely provide quantitative and qualitative differences in the respective PDFs (see Labropulu, Sullivan and Ye [7] and [8] for a discussion of this effect). The issue here is that one cannot creditably validate a closure hypothesis with the more convenient steady laboratory data and extrapolate to contaminant cloud concentration reduction. Contaminant clouds must be measured.

To make progress in the important problem of describing concentration reduction in contaminant clouds, one must have a non-subjective (see the Chatwin and Sullivan [9] discussion of an "intermittency factor" in this regard) and representative measure that can actually be determined experimentally.

2. – The expected mass fraction

The expected-mass-fraction function is simply described. Consider a contaminant cloud at time t and sum the continuum scale volume elements δv that contain a particular concentration θ. Taking the numerical average of this same quantity $\delta m(\theta) = \theta \delta v$ over many repeat cloud releases and dividing by the conserved release mass Q provides the expected mass fraction at this concentration. Thus, the distribution of the release mass over various concentration levels is portrayed as the cloud evolves in time. Mixing is evidenced by the transfer of contaminant, due to molecular diffusivity κ, to lower values of concentration.

More formally, the expected-mass-fraction function, emf, is defined as

$$(3) \qquad \tilde{p}(\theta; t) = Q^{-1} \int_{\text{a.s.}} \theta p(\theta; \mathbf{x}, t) \, \mathrm{d}v,$$

where a.s. indicates an integral over all space, with moments

$$(4) \qquad \tilde{m}_n(t) = \int_0^\infty \theta^n \tilde{p}(\theta; t) \, \mathrm{d}\theta = Q^{-1} \int_{\text{a.s.}} m_{n+1}(\mathbf{x}, t) \, \mathrm{d}v,$$

and

$$(5) \qquad \int_0^\infty \tilde{p}(\theta; t) \, \mathrm{d}\theta = 1.$$

$\int_{\theta_a}^{\theta_b} \tilde{p}(\theta; t) \, \mathrm{d}\theta$ is the fraction of release mass Q found, on average, in the cloud in the concentration interval between θ_a and θ_b at time t.

There are intuitive advantages in using 'cloud-average' statistics (see Chatwin and Sullivan [10] and Sullivan and Ye [11]). $\tilde{p}(\theta; t)$ has no spatial reference in its definition and uses all values of contaminant concentration in each realization in its compilation such that convergence with relatively few realizations is expected. The definition requires only that the contaminant be conserved. For example, it could be applied separately to describe concentration of saline solution and heat in the same experiment. The definition could be modified to apply to non-conserved contaminant, for example when chemical reactions take place, by replacing Q with $\overline{Q(t)}$ in (3) and (4).

If one were dealing with a combustible contaminant, for example, the mass of contaminant that is, on average, within the flammable limits θ_1 and θ_u at time t is

$$(6) \qquad\qquad Q \int_{\theta_1}^{\theta_u} \tilde{p}(\theta; t)\,\mathrm{d}\theta.$$

The fraction of release mass that is, on average, above a noxious or dangerous concentration level θ_1 is

$$(7) \qquad\qquad \int_{\theta_1}^{\infty} \tilde{p}(\theta; t)\,\mathrm{d}\theta.$$

There are very few published measurements on contaminant cloud concentrations and none that could be used to compute $\tilde{p}(\theta; t)$. Ye [12] describes, in detail, a feasible experiment that could be carried out (and which to some extent mitigates the severe resolution problems raised in the introduction), using an extension of the litmus-fluid experiments of Corriveau and Baines [1], to measure $\tilde{p}(\theta; t)$. Here some meandering plume data (Sullivan [13]) from the well-mixed surface layer of Lake Huron will be used to illustrate the effectiveness of the above approach.

In the experiment dyed fluid is continuously released from a small-diameter source into the surface layer of a lake. The dye plume so formed is contained above the thermocline and below the free surface. At fixed downstream distances $x = \overline{U}t$, where $\overline{U}$ is the uniform mean flow velocity, the plume is repeatedly traversed with a small boat containing a sample probe. Although the statistical properties of the plume are taken to be depth independent, the probe samples at only one fixed depth during each traverse. To reinforce the statement made in the Introduction, few repetitious plume crossings could be made (at most 25 at 2 downstream locations) over a period for which the flow conditions remained reasonably unchanged. Of course the $\tilde{p}(\theta; t)$ given in (3) is the same whether the $p(\theta; \mathbf{x}, t)$ is measured in a fixed (absolute diffusion) or center-of-mass (relative diffusion) reference frame. The conserved quantity in this experiment is the flux of contaminant across a plane,

$$(8) \qquad\qquad \int \overline{U\Gamma(\mathbf{x}, t)}\,\mathrm{d}A \doteq \overline{U} \int \overline{\Gamma(\mathbf{x}, t)}\,\mathrm{d}A,$$

where the integral is over the plume normal to the constant mean flow velocity $\overline{U}$. Ideally, one would like to use a planar version of $\tilde{p}(\theta; t)$ where a concentration record over the entire plume is available rather than the record over one line through the plume in each realization, as is the case in these experiments. Inevitably there is a considerable variation in the total contaminant mass recorded in the individual traverses in these experiments.

In fig. 1 the $p'(\theta)$ is shown for 20 plume crossings at one downstream sampling station. The $p'(\theta)$ are compiled by accumulating all of the contaminant in a given concentration interval $\Delta\theta$ (in arbitrary concentration units) from the digitized concentration record,

$$(9) \qquad\qquad r_j(\theta) = \sum_i \Gamma_i\,\Delta t \quad \text{for} \quad \theta < \Gamma_i < \theta + \Delta\theta$$

and normalizing such that

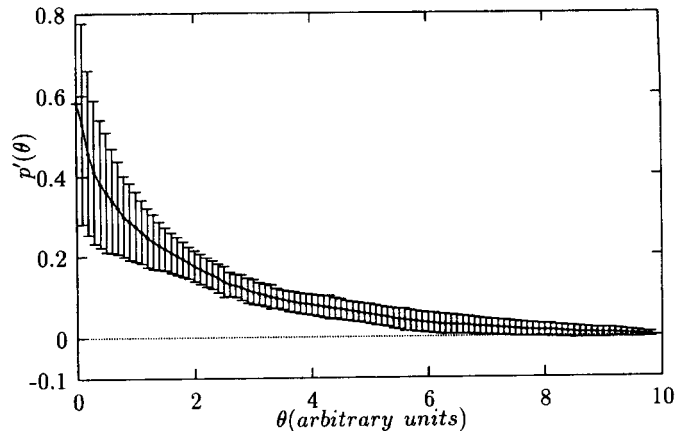$$(10) \qquad\qquad p'(\theta) = r_j(\theta) \Big/ \sum_j r_j(\theta)\,.$$

Fig. 1. – The expected-mass-fraction function for Experiment 1, Section 1 of Lake Huron plume data.

The shape of $p'(\theta)$ appeared to be not much changed from realization to realization (see Ye [12]). The root-mean-square bars included in fig. 1 indicate a relatively small quantitative variation from realization to realization and this variation would be expected to be much less if concentration values over the entire sample plume cross-section were incorporated. Similar results were found at other sample stations.

The variability of $p'(\theta)$ must be assessed in the context of any alternative representation of contaminant dilution. The experiments were originally conducted to assess mean concentration values in center-of-mass coordinates. There are insufficient realizations to adequately measure the distributed second moment as shown in Chatwin and Sullivan [14]. It would be out of the question to attempt to compile, for example, the probability density function, even in center-of-mass coordinates.

Experimental data on contaminant clouds is usually in the form of a concentration record taken as the cloud passes over a fixed sampling position, say $\mathbf{x}_0$. Heagy and Sullivan [15] have developed a fixed-point version of the emf and used this to analyze the Hall *et al.* [6] data. The fixed-point emf is defined by

$$(11) \qquad \hat{p}(\theta; \mathbf{x}_0) = \overline{d}^{\,-1}(\mathbf{x}_0) \int_0^\infty \theta p(\theta; \mathbf{x}_0, t)\, \mathrm{d}t,$$

with moments

$$(12) \qquad \hat{m}_n(\mathbf{x}_0) = \int_0^\infty \theta^n \hat{p}(\theta; \mathbf{x}_0)\, \mathrm{d}\theta = \overline{d}^{\,-1}(\mathbf{x}_0) \int_0^\infty m_{n+1}(\mathbf{x}_0, t)\, \mathrm{d}t$$

and

$$(13) \qquad \int_0^\infty \hat{p}(\theta; \mathbf{x}_0)\, \mathrm{d}\theta = 1.$$

The value of $\overline{d}(\mathbf{x}_0)$ is found from taking the ensemble average of the random variable "dosage",

$$(14) \qquad d(\mathbf{x}_0) = \int_0^\infty \Gamma(\mathbf{x}_0, t)\, \mathrm{d}t,$$

as

$$(15) \qquad \overline{d}(\mathbf{x}_0) = \int_0^\infty \overline{\Gamma(\mathbf{x}_0, t)} \, \mathrm{d}t = \int_0^\infty m_1(\mathbf{x}_0, t) \, \mathrm{d}t \,.$$

A discussion of the variability of $\hat{p}(\theta; \mathbf{x}_0)$ from realization to realization is given and that variability is shown to be comparable with the variability of the (low information statistic) dosage. It is worth mentioning that $\hat{p}(\theta; \mathbf{x}_0)$ can also (instead of using a direct binning procedure) be easily approximated from experimental data by simply measuring

$$(16) \qquad \overline{\int_0^\infty \Gamma^n(\mathbf{x}_0, t) \, \mathrm{d}t}$$

for some few small values of n (say $n \leq 5$) and, from (12), use these in an inversion procedure, as mentioned in the Introduction, to arrive at $\hat{p}(\theta; \mathbf{x}_0)$.

The expected mass fraction provides a view of the state of a cloud as frozen at a fixed instant in time, whereas the fixed point version records the state as the evolving cloud is convected past the fixed point. The important point to be made here with respect to experimental validation is that the fixed point version of the emf converges with sufficiently few realizations that it is practical to measure it in a laboratory experiment such as that of Hall *et al.* [6]. For example, predictions of $m_2(\mathbf{x}_0, t)$, the emf and the fixed point emf for clouds released in the neutral atmospheric boundary layer are given in Labropulu and Sullivan [16], Sullivan and Ye [17] and Heagy and Sullivan [18], respectively. The fixed point emf and $m_2(\mathbf{x}_0, t)$ could be measured, for example with some slight modifications to the experimental design of the Hall *et al.* [6] experiments, and direct inferences made about the validation of the emf.

3. – Moments

Chatwin and Sullivan [10] provide an exact and rather general equation for the moments

$$(17) \qquad \frac{\mathrm{d}}{\mathrm{d}t} \int_{\mathrm{a.s.}} \overline{\Gamma^{n+1}(\mathbf{x}, t)} \, \mathrm{d}v = -n(n+1)\kappa \int_{\mathrm{a.s.}} \overline{\Gamma^{n-1}(\mathbf{x}, t)(\nabla\Gamma(\mathbf{x}, t))^2 \, \mathrm{d}v},$$

which, using the definition of (12), provides

$$(18) \qquad \tilde{m}_n(t) = -n(n+1)\kappa \int_0^t f(t') \, \mathrm{d}t' + \tilde{m}_n(0),$$

where

$$(19) \qquad f(t) = Q^{-1} \int_{\mathrm{a.s.}} \overline{\Gamma^{n-1}(\mathbf{x}, t)(\nabla\Gamma(\mathbf{x}, t))^2 \, \mathrm{d}v} \,.$$

The idea is that if all of the moments are known then, generally, one can reproduce $\tilde{p}(\theta; t)$ and one expects to get a good approximation from the first few low-order moments $\tilde{m}_n(t)$. One apparent advantage is the relatively simple form of eqs. (17) and (18) and the prospect of affecting closure on the $(\nabla\Gamma)^2$ term that appears in the right-hand side of (17) under an integral over all space. This has been shown to have promise, in conjunction with the simple moment prescription given in Chatwin and Sullivan [19], in Labropulu and Sullivan [16] and Clarke and Mole [20] and will not be pursued further here.

It is of interest to consider the implications of the sheet-strand texture of the contaminant distribution mentioned in the Introduction. Specifically, that virtually all of the
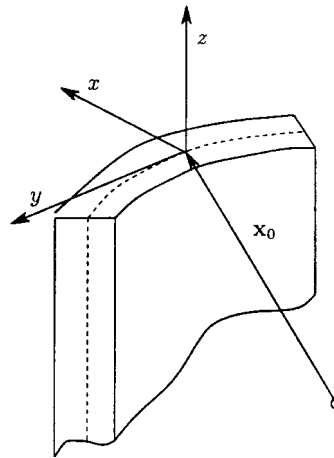
Fig. 2. – A sketch of a patch of an isolated contaminant sheet. The x axis is normal to the sheet and the (y, z) plane is tangent to the sheet. The thickness is comparable with the Batchelor conduction cut-off length.

contaminant was found to be in sheets of a thickness comparable with the conduction cut-off length λ by Corriveau and Baines [1]. The fine-scale inhomogeneity is seen to be more pronounced in the observations of $(\nabla\Gamma)^2$ in Dahm *et al.* [2] where it also appears that the sheet separation is much larger than the sheet thickness.

Consider the isolated segment of a contaminant sheet sketched in fig. 2. The only significant gradients are taken to be those in the direction x across the sheet. It will be assumed that the concentration distribution across a sheet will be of the self-similar form

$$(20) \qquad \Gamma(x) = \Gamma_0 F(\eta) \,; \quad \eta = x/\overline{\lambda} \ .$$

This self-similarity presumes that the straining-diffusive mechanism that generates the sheets is very much the same for all sheets. Γ_0 is taken to be a representative, say local sheet maximum, concentration and $\overline{\lambda}$ is taken to be a length scale that is approximately λ throughout the cloud at a given time. Relevant terms that appear in (17) become, in each realization,

$$(21) \qquad \int_{\text{a.s.}} \Gamma^{n+1}(\mathbf{x}, t)\, \mathrm{d}v = \overline{\lambda}(t)a(n)R(t) \,,$$

$$(22) \qquad \int_{\text{a.s.}} \Gamma^{n-1}(\mathbf{x}, t)(\nabla\Gamma(\mathbf{x}, t))^2\, \mathrm{d}v = \frac{b(n)R(t)}{\overline{\lambda}(t)} \,,$$

$$(23) \qquad R(t) = \int_s \Gamma_0^{n+1}\, \mathrm{d}\sigma \,,$$

where the integral is over all of the sheets surface area.

$$(24) \qquad b(n) = \int_{-\infty}^{\infty} F^{n+1}\left(\frac{F'}{F}\right)^2 \mathrm{d}\eta \,,$$
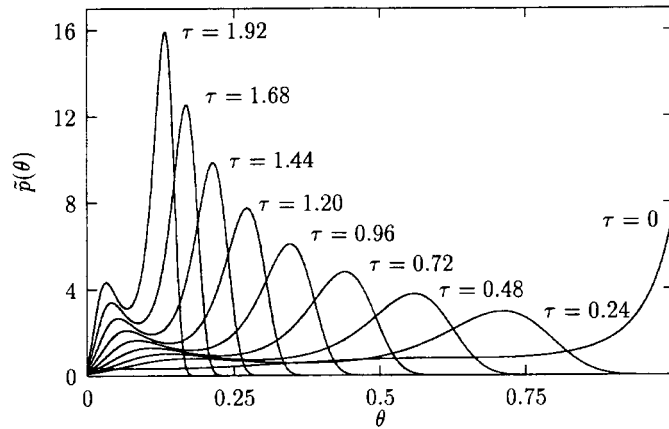
Fig. 3. – The expected-mass-fraction function for a cloud with a Gaussian-sheet texture.

$$(25) \qquad\qquad a(n) = \int_{-\infty}^{\infty} F^{n+1} \, \mathrm{d}\eta \,.$$

This provides the equation, after taking an ensemble average,

$$(26) \qquad\qquad \frac{\mathrm{d}}{\mathrm{d}t} (\overline{\overline{\lambda} a R}) = \frac{-\kappa n(n+1)}{\overline{\lambda}^2} \left(\frac{b}{a} \right) (\overline{\overline{\lambda} a R}) \,,$$

which, using (12), provides the solution

$$(27) \qquad\qquad \tilde{m}_n(t) = \tilde{m}_n(0) \exp \left[-\kappa n(n+1) \left(\frac{b}{a} \right) \int_0^t \frac{\mathrm{d}t'}{\overline{\lambda}(t')^2} \right] \,.$$

To be more specific, for the purpose of illustration, if the self-similar concentration is taken to be the Gaussian form

$$(28) \qquad\qquad \Gamma(x) = \Gamma_0 \exp \left[\frac{-(x - x_0)^2}{2\overline{\lambda}^2} \right] \,,$$

then

$$(29) \qquad\qquad \frac{b(n)}{a(n)} = \frac{1}{n+1} \,.$$

In situations where $\overline{\lambda}$ is independent of t, for example a cloud dispersing in the inertial subrange or in steady homogeneous turbulence, the solution (27) becomes

$$(30) \qquad\qquad \tilde{m}_n(\tau) = \tilde{m}_n(0) e^{-n\tau}; \quad \tau = \frac{t\kappa}{\overline{\lambda}^2} \,.$$

It is shown in Ye [12] that the moments provided by (30) satisfy the conditions required for a unique representation, through inversion, of $\tilde{p}(\theta, \tau)$. $\tilde{m}_n(0) = \theta_0^n / \sqrt{n+1}$ in (30). The $\tilde{p}(\theta, \tau)$ shown in fig. 3 are compiled by setting $\theta_0 = 1$ and using the lowest four moments from (30) with the maximum entropy method given in Derksen and Sullivan [4].
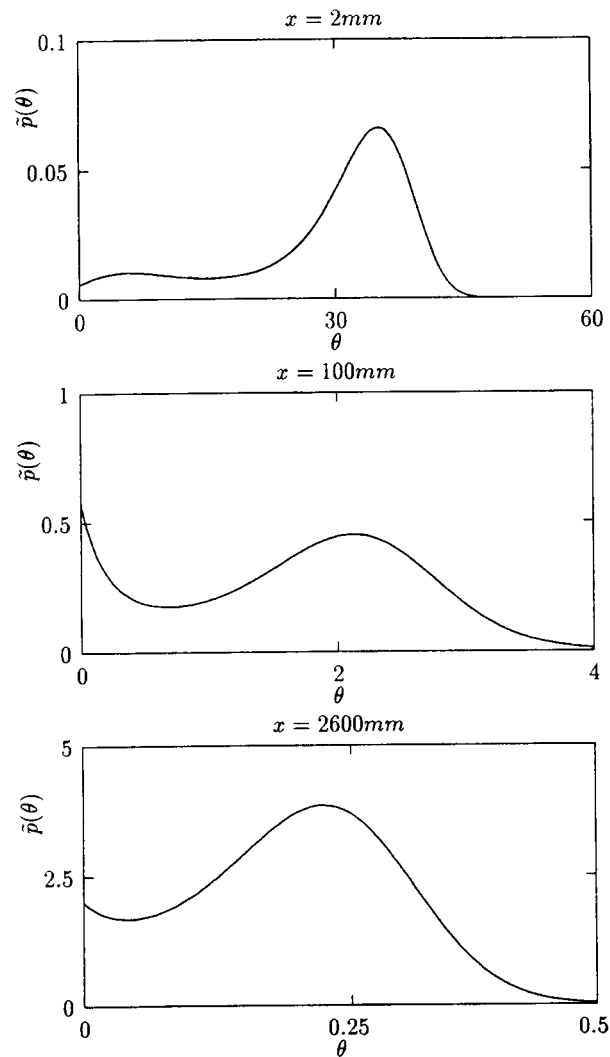
Fig. 4. – The expected-mass-fraction function for a steady plume in grid-turbulence at three down-stream locations.

Figure 3 clearly shows the migration of mass to lower concentration values as τ increases. At $\tau = 0$ one does not observe a delta function at $\theta/\theta_0 = 1$ since, physically, the process requires a finite 'fragmentation' time for the sheet-strand structure to develop. The bimodal shape that appears at large τ could be an anomaly associated with the inversion procedure, however this feature does appear in some measurements to be discussed presently.

The adequacy of the above, simple, presentation of the emf can only be determined by experiment. There are no suitable cloud data available, however, a planar version for use with the steady-source, uniform flow, homogeneous, wind-tunnel, grid turbulence experimental data can be developed. Equation (17) with downstream distance $x = \overline{U}t$, where the integrals and gradients are taken to be over planes normal to the mean velocity

$\overline{U}$ and mixing in the streamwise direction is neglected, becomes

$$(31) \qquad \overline{U}\frac{\mathrm{d}}{\mathrm{d}x}\int_A \overline{\Gamma^{n+1}}\,\mathrm{d}A = -\kappa n(n+1)\int_A \overline{\Gamma^{n-1}(\nabla\Gamma)^2}\,\mathrm{d}A \ \ .$$

The conserved quantity is the flux as given in (8) and, provided the variation of λ in the streamwise direction is neglected, the solutions are as given in (30) with t replaced by $x/\overline{U}$.

In fig. 4 the planar version of the emf is compiled for the plume generated by a heated wire, steady, line-source in the grid turbulence measurements of Sawford and Sullivan [22]. Measured moments at fixed locations across the plume are used to compile the planar emf at three downstream locations. The basic shapes shown in fig. 4 are qualitatively similar to those of fig. 3 and the migration of mass to lower concentration values at further downstream locations is clearly in evidence. To compile the integrated moments, and hence the smooth emfs shown in fig. 4, the Chatwin and Sullivan [19] moment prescription, which was shown to be a very good representation of the experimental data, was utilized. The procedure is discussed in Ye [12]. A discussion of measured, low-concentration, values is difficult due to an inevitable 'thresholding' factor (see Lewis and Chatwin [21]). It would be very instructive to compare data from experiments specifically targeted to measure the spatially integrated concentration moments. Measurements, for example, taken in the approximately homogeneous central region of a large diameter, high Reynolds number, pipe flow (where λ is constant) and where appropriate measures are taken to account for thresholding effects, would be especially enlightening.

4. – Concluding remarks

It was shown in the Introduction that conventional measures of contaminant mixing in clouds are not directly practical or adequate due to the problem of taking averages and the essential requirement that all approximations must be experimentally validated. A new measure, the expected-mass-fraction function, is introduced. This new function has a clear and immediate interpretation and the (fixed-point version of) emf can be measured, in that reasonable ensemble averages can be approximated from conventional laboratory experiments. Ideally, one would like a joint multipoint probability density function to describe turbulent mixing in contaminant clouds. The emf is a more modest measure that can be derived from approximations on a relatively simple differential equation. This degree of simplicity is consistent with the information that would be available on release and flow conditions in describing the accidental spill of contaminant into an environmental flow. Reasonable, physical, approximations are shown to lead to qualitatively sensible forms of the emf, however, further progress is inhibited by the need for specifically targeted detailed experimental data.

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