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MIXING: KINETICS AND GEOMETRY

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Abstract We review the different facets of the phenomenon of mixing, including its geometrical, temporal, and structural aspects. Then we suggest that a complex mixture can be viewed as the superposition of independent sources. Kinetics and geometry are shown to be closely linked to each other when following the transient mixing of an isolated scalar source in a turbulent flow. The composition law between multiple interacting sources is established experimentally, therefore allowing one to reconstruct any scalar field from well defined elementary contributions.

1. WHY MIXING?

Mixing¹ is a subject that suffers from the *Bourgeois Gentilhomme* complex. Like Monsieur Jourdain in Molière’s play (1670), scientists, engineers, and indeed all of us often “do mixing without even knowing it” (just think about yourself trying to prepare mayonnaise ...).

As an operation, it consists simply in putting together two or more initially segregated constituents and stirring, in order to attain uniformity, or a new product, or the complete disappearance of one of the constituents, etc.; mixing thus is indeed at the crossroads of many different areas of science. One often needs to mix elements in order to make a new product, a homogeneous blend, or to make possible a chemical reaction or an efficient combustion. One also needs to understand how nature mixes or has mixed to gain information on, e.g., the size of a pollutant spot in a valley, the rate of destruction of ozone in the atmosphere, or the dynamics of the earth’s mantle. More than a fascinating subject in itself, mixing is thus ubiquitous and a key process in many complex man-made or natural operations.

¹The text in this section is inspired from the foreword of *Mixing: Chaos and Turbulence*, Chaté, H., E. Villermaux, and J. M. Chomaz (eds.), Kluwer Academic/Plenum Publishers, New York, 1999.

It is no doubt *because* of its universality that mixing, as noted with regret by J. Ottino (1989), “... does not enjoy the reputation of being a very scientific subject ...” as is frequently the fate of interdisciplinary topics.

The dendritic nature of the subject matter is revealed by the different angles from which the various scientific communities attack the problem, according to their needs.

These approaches can be roughly grouped into three naturally overlapping categories, each of them of interest to different schools of thought:

- Geometry
- Kinetics
- Structures

Beyond the tools and attitudes developed by different scientific groups regarding the problem of mixing, one might appreciate that mixing, as suggested by common sense, is the operation by which a system evolves from one state of simplicity (the initial segregation) to another state of simplicity (the complete uniformity). Between these two extremes, complex patterns emerge and die. Questions then naturally arise: how can the geometry of complex patterns be characterized; what is the clock, the time-scale of the process; and what are the structures involved in the flow?

1.1. Geometry

Very early on, emphasis was placed on the geometry of the mixing zone in the combustion context. Indeed the involved, multiscale geometry of the interface that separates two streams being mixed is not only a spectacular facet of the process, but also sometimes at the core of the physical problem. Exothermic reactions in gases, or reactions with a fast kinetics in liquids confine the reaction zone to a thin region of space and the flame appears as a sharp interface for most scales in the flow. The total extent of the flame area dictates the propagation speed of the front in premixed reactants, as noted by Damköhler (1940), and the net combustion rate in diffusion flames (see the celebrated experiments of Hawthorne et al. (1949)). Knowing how and why the front is distorted by the underlying motions is thus crucial for predicting the flame extent.

The multiscale structure of the contour of a scalar blob immersed in a disordered flow was recognized by Welander (1955), who suggested how a connection could be made between the internal structure of the underlying motions and the complexity of the blob shape. Welander even

made reference to the Koch curve, long before fractals were popularized in this and other contexts by Mandelbrot (1975).

The program, still very active today, for investigating the geometry of the scalar support, or the iso-concentration surfaces in flows was certainly already contained in Reynolds (1894). Reynolds was probably even more ambitious, suggesting that watching the dynamics of “coloured bands” in a flow was a route towards understanding the driving motions.

1.2. Kinetics

Mixing is, in the strict sense, a transient process from initial segregation to ultimate homogeneity. This calls for the understanding of the kinetics and timescales of the process.

Studies on dispersion address the problem of the growth rate of the radius of a tracer blob in a prescribed displacement field. In addition to pure molecular diffusion in the medium at rest, fluid motion usually enhances dispersion (Taylor 1921, 1953) and alters not only the diffusion law, by a renormalization of the diffusion coefficient, but also the structure of the law itself. Due to persistent ballistic movements and ever larger jumps in turbulent flows, dispersion laws exhibit, in the absence of traps or slow recirculating motions, a faster-than-linear growth of the mean-squared radius of the blob (Prandtl 1925, Richardson 1926). The presence of bypasses or dead-ends in complex geometries alter, in continuous flow systems (a river, a valley through which wind blows, an open chemical reactor), the residence time distribution of a tracer introduced at the inlet of the system. The novel features of the distribution are spikes at short times if a short circuit is present, and/or long tails caused by traps and slow motions in confined cavities. This was first described by Danckwerts (1953).

The kinetics of mixing does not only refer to dispersion. Dispersion may result solely from a spatial reorganization of the quantity to be mixed, with no interpenetration with the substrate at the molecular level.

Mixing, as opposed to stirring, actually means homogenization at the smallest, i.e. diffusive, scales; and the appropriate quantity to define and measure the mixing time, namely the “intensity of segregation,” was introduced by Danckwerts (1952). The mixing time (for instance in a tank stirred with an impeller, familiar in the chemical industry) is found to be solely determined by the large-scale features of the flow (integral scale, root-mean-square velocity; see, e.g., Nagata (1975)), regardless of the intimate structure of turbulence, provided the Reynolds number is

large enough (typically larger than 10^4), in a wide variety of flows. This absence of a role for turbulence and its structure in the characteristic time of the inhomogeneities decay is an important point, which has been somewhat overlooked. The intimate structure of the local rearrangements in the flow does, however, play a role when chemical reactions with a nonlinear kinetics, or consecutive-concurrent chemical reactions occur within the mixture (Danckwerts 1953, Epstein 1990).

1.3. Structures

Disordered flows develop a broad hierarchy of scales of motion that convect and distort the scalar field. Notwithstanding the fact that mixing is, like the decay of the energy-containing eddies, a transient phenomenon, the problem of the interaction of the scalar field with the underlying turbulent flow has focused on hypothetical “stationary conditions.” This approach parallels the Kolmogorov quasi-equilibrium picture of turbulence (Corrsin 1951, Oboukhov 1949, Batchelor 1959, Batchelor et al. 1959) and it is in this assumed limit that the timescales of the stirring motions that distort the scalar field are all shorter than the global mixing time (i.e. the variance of the scalar fluctuations is stationary as seen on the timescale of these motions), allowing the possibility of resorting to cascade arguments, and spectral analysis.

It is customary, in turbulence, to oppose the statistical approaches, the cascade representation being one of those, to the approaches focusing on “structures” more or less coherent or permanent in the flow. This opposition is somewhat artificial. Modern statistical models of turbulence rely in fact on a description of the flow as a hierarchical structure in the real space, possibly fractal, the structure of the hierarchy inducing the statistics. The cascade picture, notably, is thus in a sense a “structural approach.”

In this cascade picture, the mixing time is the time needed for the scalar to travel, by a process of successive reductions of scale imposed by the hierarchy of pre-existing scales of motion (eddies) in the flow, from its initial size to the dissipation scale, prescribed by the Reynolds and Schmidt numbers (Corrsin 1964). This description presents many shortcomings, the first one being its inconsistency with the observed fact that the mixing time depends linearly on the initial size of the blob to be mixed, instead of on its size raised to the power $2/3$. If it is clear that mixing macroscopic objects implies a reduction of their transverse size and a multiplication of ever smaller scales for diffusion to act efficiently, there is no direct proof that a scalar blob should follow the “Kolmogorov cascade” *step by step* to be ultimately erased by molecular

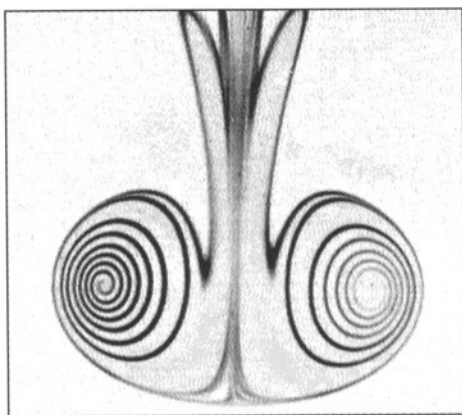


Figure 1 Start-up vortex. A fast chemical reaction between the ring and the ambient fluid traces the spiraling interface between the two media.

diffusion. Here is a sign that, in turbulence, even many basic properties escape our precise understanding; the future should therefore be rich in forthcoming works (see the program reviewed by Shraiman and Siggia 2000).

The will to reduce a complicated problem such as turbulence to a set of elementary objects containing, presumably, all of the desired information, is in keeping with a long tradition in physics and fluid mechanics. Coherent structures, filaments, worms, and sheets are frequently-invoked paradigms. In the context of mixing, the discovery of large-scale vortical structures sustained by shear flows has prompted a surge of activity (Brown and Roshko 1974). Originating essentially from a Kelvin–Helmholtz type of instability, these intense, long-lived structures are serious candidates to compete with the cascade representation since they directly couple injection scales with dissipation scales, eliminating intermediary steps. Their existence alone is not, however, sufficient to ensure a rapid mixing. When formed by a given velocity difference across a shear layer, their size (equivalent to the Reynolds number) has to grow beyond a critical value to allow the onset of the fine-scales activity, thereby hastening the uniformization within the layer. This is the so-called “mixing transition” (Breidenthal 1981).

Although its ubiquitous character is striking and goes far beyond the context of mixing layers, as recently reviewed by Dimotakis (2000), little is known in detail about what occurs during and after this transition. It is, nevertheless, a known fact that the deformation tensor in turbulent flows bears, in the mean, two directions of stretching and one direction of compression as first analyzed by Betchov (1956). This property

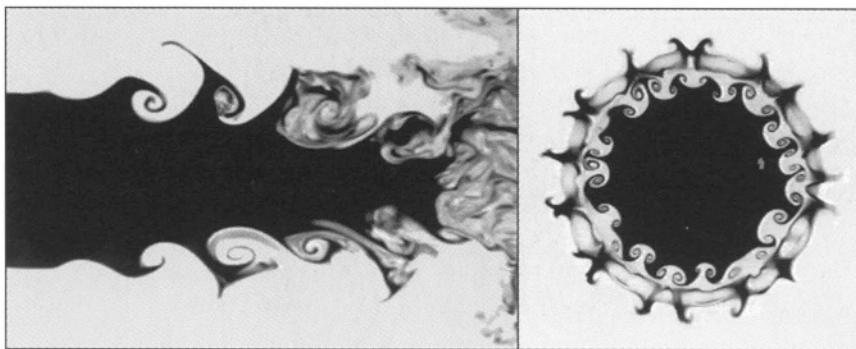


Figure 2 Cuts in the longitudinal and transverse directions of the flow of a coaxial jet showing the rolling-up structures resulting from the shear instability between the streams (from Villermaux and Rehab 2000).

leads to an increase in the length and area of material lines and surfaces (Batchelor and Townsend 1956). The stretched sheet is thus likely to be the elementary brick of mixing, whether turbulent or not (Ranz 1979, Ottino 1989).

1.4. Towards a new paradigm

Restricting our field of sight to the particular limit of mixing in homogeneous fluids, and keeping the Pandora's box of mixing in complex, non-Newtonian fluids, multiphase flows, granular flows, etc., carefully sealed, we may wonder what the minimal set of ingredients should be to reach a satisfactory description of the above three facets of the mixing phenomenon. Let us examine mixing patterns obtained in different flow conditions, going from laminar (Fig. 1) to strongly turbulent (Fig. 3), via transitional shear instabilities (Fig. 2).

Figure 1 shows a start-up vortex (a kind of a smoke ring), several turns after its formation. The fluid constitutive of the ring develops a chemical reaction with the ambient medium in which it is rolled up. This is a fast chemical reaction and the reaction diffusion zone, very thin in this case, traces the spiraling interface between the two media. The ring is a non-turbulent object. It directly couples its own injection scale with the diffusive scale via the spiraling motion and the accretion of sheets of ambient fluid at each turn. However, the ring presents many scales, even a continuous spectrum of scales, due to the continuous increase of the striation thickness from the core of the spiral to its edge. This is a fractal whose covering dimension is close to $3/2$. This is a multiscale, dissipative object; but where is the cascade?

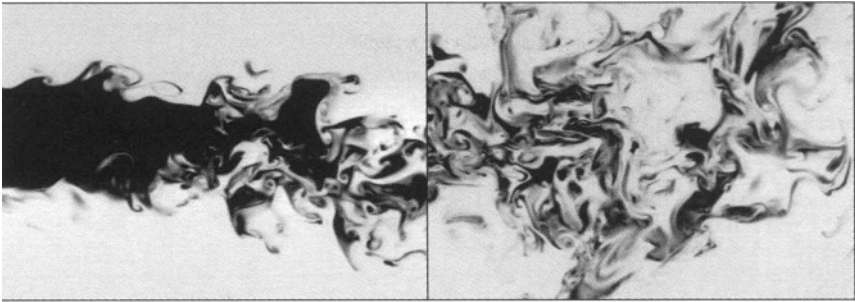


Figure 3 Blob of dye deposited in a large scale sustained turbulent flow converted into disjointed sheets which dilute in the surrounding medium (from Villerraux and Innocenti 1999).

Figure 2 represents cuts in the longitudinal and transverse directions of the flow of a coaxial jet. The inner jet is slower than the outer annular jet, and the pictures show the structures that result from the instability of the shear between the streams. One recognizes in the longitudinal structures formed from the shear instability the rolling-up vortices similar to those in Fig. 1. But the coherence of these structures gets lost and the resulting mixture finally looks like Fig. 3.

Figure 3 illustrates what happened to a small compact blob of scalar deposited in a large-scale sustained turbulent flow. The blob has been progressively converted into disjointed sheets that dilute in the surrounding turbulent medium. Except for loose, diffuse sheets of dye, no obvious structures can be distinguished. The striking observation is nevertheless the broad fluctuations in the transverse size of the sheets, and in the concentration level they carry. Although captured at the same instant of time in the snapshot of Fig. 3, those sheets have not all experienced the same history. Some are still thick and dark, while others are already so thin that they almost fade away in the diluting medium.

What is the paradigm, the elementary structure of mixing in disordered flows? The coherent rolling-up vortex, the cascade, the stretched sheet? It seems that none of these caricatures is fully satisfactory and could give way to a new paradigm that would account for the parallel, distributed histories of cumulated stretchings experienced by the material elements in the course of time. This new paradigm could reconcile the three facets of mixing by unifying geometrical, temporal, and structural aspects of the process: its intrinsic transient nature, its evolutive geometry as a set of particular structures, namely sheets with distributed histories.

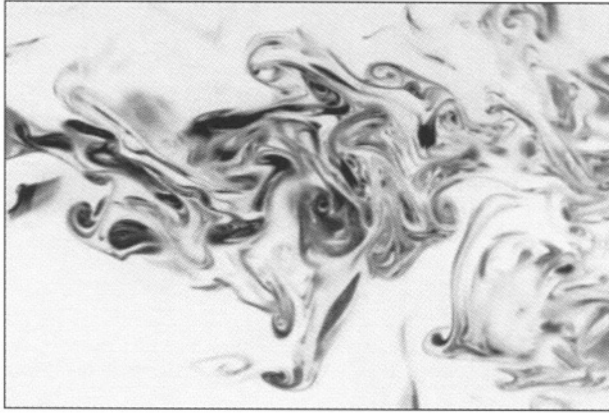


Figure 4 A snapshot of the scalar distribution in the region $8 < x/d < 12$ downstream of the injection point, with $d = 1$ cm and $Re = u' L / \nu = 6000$.

2. FROM ELEMENTARY SOURCES TO COMPLEX MIXTURES

2.1. The single source

Experiments (Villermaux et al. 1998, Villermaux and Innocenti 1999) have been conducted aiming at following an initially smooth and compact blob of dye released in a turbulent flow along its transient evolution, from the initial segregation, towards uniformity. The scalar to be mixed is injected continuously in the far field and on the axis of a turbulent jet via a small tube whose diameter d is smaller than the local integral scale L (typically $d/L \approx 1/10$). The injection point behaves neither as a source nor as a sink of momentum, in the mean, and the properties of the flow (stirring scale L , r.m.s. velocity u') are constant during the uniformization period of the scalar.

These experiments involve three types of scalars: temperature in air ($Sc = \nu/D = 0.7$), temperature in water ($Sc = 7$), and the concentration of disodium fluorescein in water ($Sc = 2000$), thus allowing an investigation of the quantitative role of the intrinsic diffusive molecular properties of the scalar being mixed on the process.

Mixing time. The blob is progressively converted into a set of stretched sheets, possibly coalescing as they fade away (Fig. 4), and the scalar concentration fluctuations PDF, $P(C/C_0)$, where C_0 denotes the injection concentration, exhibits rapidly a self-preserving shape, whose

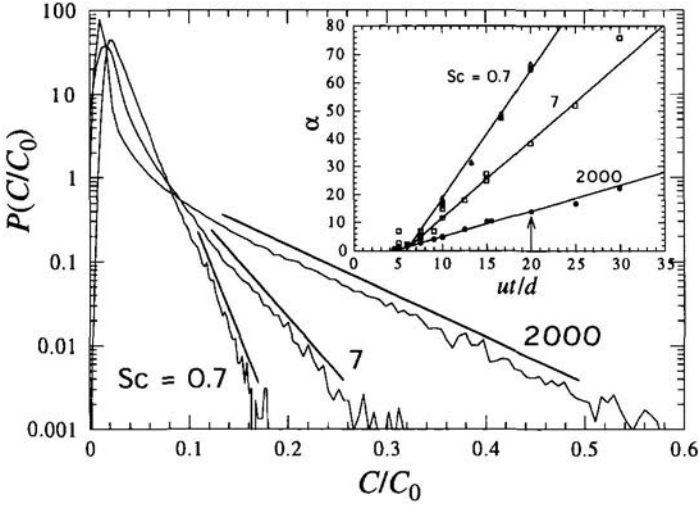


Figure 5 Fluctuations PDFs, normalized by the initial concentration (temperature), recorded for three Schmidt numbers 20 diameters d downstream from the injection point. The PDFs exhibit an exponential decay of the form $P(C/C_0) \sim \exp(-\alpha C/C_0)$. Insert: The argument of the exponentials at different times ut/d and three Schmidt numbers. \bullet $Sc = 2000$, $Re = 6000$ and 12000 , $d/L = 0.05, 0.1, 0.6$. \square $Sc = 7$, $Re = 6000$, $d/L = 0.05, 0.1, 0.16$. \blacksquare $Sc = 0.7$, $d/L = 0.08$, $Re = 23000$; \circ, Δ $Sc = 0.7$, $d/L = 0.08$, $Re = 45000$.

tail is an exponential with an argument increasing linearly in time t as

$$P\left(\frac{C}{C_0}\right) \sim \exp\left(-\frac{t}{t_s} \frac{C}{C_0}\right), \quad \text{for } t \gg t_s, \quad (1)$$

with the mixing time t_s being found to be

$$t_s = \frac{d}{u'} f(Sc), \quad (2)$$

as shown in Fig. 5. The factor $f(Sc)$ is a slowly increasing function of the Schmidt number. A fit consistent with the data is $f(Sc) \sim \ln(Sc)$, although a weak power-law dependence of the form $f(Sc) \sim Sc^{0.2}$ is not inconsistent as well.

The use of three different injection diameters indicates that the mixing time of a blob of size d scales like $t_s \sim (d/u') \ln(5Sc)$, as opposed to $(L/u')(d/L)^{2/3}$ according to the vision proposed by Corrsin (1964) and Oboukhov (1949). The spectrum of the mixture is also found to decay like k^{-1} , in the inertial range of scales, and for scales smaller than the injection scale d , as opposed to $k^{-5/3}$. Moreover, the mixing time t_s

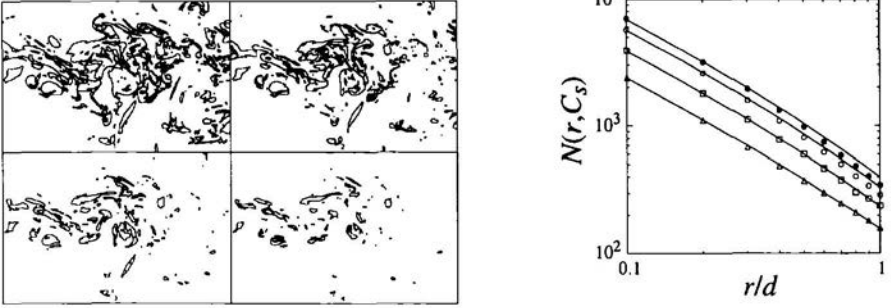


Figure 6 Left: Contours of the level sets of the scalar field of Fig. 4, for four different concentration thresholds levels. From left to right and top to bottom, $C_s/C_0 = 0.2$, 0.3 , 0.4 and 0.5 . Right: Corresponding box-counting relationships. $\bullet C_s/C_0 = 0.2$, $\circ 0.3$, $\square 0.4$, $\triangle 0.5$. Continuous lines fit by Eqn. (5).

has been found to be *smaller* than the cascade time $(L/u')(d/L)^{2/3}$. We have suggested (Villermux et al. 2000) that the process of mixing *does not* follow the sequential route expected from cascade arguments, but is on the contrary “bypassed” by a strong and constant stretching rate acting at the injection scale.

Transient geometry. The changes in the blob morphology in the course of its dilution reflect closely the uniformization process itself. This is best illustrated by following the transient shape of a particular isoconcentration contour C_s . Figure 6 shows four contours corresponding to four different concentration levels C_s .

Consider a blob of dye initially smooth and segregated. The number of segments, or square boxes of linear size r needed to cover its contour at a threshold level C_s is $N(r, t = 0, C_s) \sim 1/r$ on a two-dimensional cut such as those of Figs. 3 or 4. If the blob is immersed in a prescribed displacement field whose stationary velocity increments $\delta u(r)$ give rise to a scale-dependent stretching rate $\gamma(r) \sim \delta u(r)/r$, then the number of segments constitutive of the contour at time t is

$$N(r, t, C_s) = N(r, 0, C_s) \left[1 + \gamma(r)t \exp \left(-\frac{t}{t_s} \frac{C_s}{C_0} \right) \right], \quad (3)$$

where the amplification factor $\gamma(r)t$ expresses the net increase in the contour corrugations length, all the more rapid that $\gamma(r)$ is intense, and where the exponential factor reflects the disappearance of the scalar by molecular diffusion according to the global temporal evolution of the concentration histogram (Eqn. (1)).

The maximal rate of shear, giving the separation rate of two material points initially close to each other, is of the order of u'/d as suggested by the scale dependence of the mixing time t_s in Eqn. (2). Considering two material points belonging, for instance, to the same isoconcentration contour, initially separated by a distance r not necessarily infinitely small, the rate at which their separation distance increases $\gamma(r)$ is smaller than u'/d . Velocity gradients $\delta u(r)/r$ in the flow tend to vanish, in the mean, for separation distances r larger than the scale of the mean gradient support, namely the injection size d in the present case. A possible model for the elongation rate $\gamma(r) = \delta u(r)/r$ for any scale r is

$$\gamma(r) = \frac{\delta u(r)}{r} = \frac{u'}{d} \frac{1}{1 + r/d}. \quad (4)$$

The number of boxes are represented, for the corresponding threshold levels by (see Fig. 6)

$$N(r, t, C_s) \sim \frac{1}{r/d} \left[1 + \frac{B}{1 + r/d} \right] \quad \text{with} \quad B = \frac{u't}{d} \exp \left(-\frac{t}{t_s} \frac{C_s}{C_0} \right), \quad (5)$$

according to (3) and (4). Accounting for the initial smoothness of the contour, namely $N(r, 0, C_s) \sim r^{-1}$, the covering relationship (5) is a combination of the trivial $1/r$ factor, times a corrective factor, increasing in magnitude with time, and whose weight depends on scale: it is, at a given instant of time, a decreasing function of scale, expressing the fact that small scales have, in proportion, more contributed to the corrugation of the contour than larger scales, precisely because shearing motions are less efficient at large scales than at smaller ones (Eqn. (4)).

The covering relationship (5) thus exhibits a curvature, whose direct consequence is the scale dependence of the fractal dimension of the contour $d_f = -d \ln N(r, t, C_s) / d \ln(r)$. This fact has been recognized in a number of related instances (see Villiermaux and Innocenti 1999 and references therein) where the local (in scale) fractal dimension is found to increase from 1 at small scale, to larger values, possibly reaching 2, characteristic of space-filling objects in two dimensions.

The above scenario provides a mechanism for this continuous transition, its origin lying in the close interplay between kinetics and geometry.

2.2. Multiple sources

A complex mixture in the real world is obviously the result of the superposition of many different sources. If we think of a hot jet discharging in a quiescent cold environment, the temperature fluctuations on its centerline at a given distance from the nozzle are certainly likely



Figure 7 Two nearby sources discharging in a turbulent medium in which they mix.

to be the result of cumulated contributions from different parcels of cold fluid entrained at the boundary of the jet in the course of its development.

The “signature” of one scalar source presented in the previous section is likely to be altered if another source is present in its vicinity (as shown in Fig. 7), and the interaction between those two has to be considered.

If $P_1(C)$ and $P_2(C)$ are the bare PDFs of each of the two sources disposed in the vicinity of each other, the compound PDF $P_{1+2}(C)$ is shown in Fig. 8.

As long as the plumes emanating from each of the sources do not interfere, the sources develop in an anticorrelated manner: the concentration measurement point is either in one plume, or in the other (see also Warhaft 1984). Then, as soon as the plumes merge, it is observed experimentally that $P_{1+2}(C)$ is very close to the *convolution* of $P_1(C)$ and $P_2(C)$. The concentration C in the resulting mixture is the sum of the concentration C_1 from source 1 and of concentration C_2 from source 2, C_1 and C_2 being chosen independently in each of the original distributions provided that

$$P_{1+2}(C) = \int_0^C P_1(C_1)P_2(C_2)dC_1, \quad \text{with } C = C_1 + C_2. \quad (6)$$

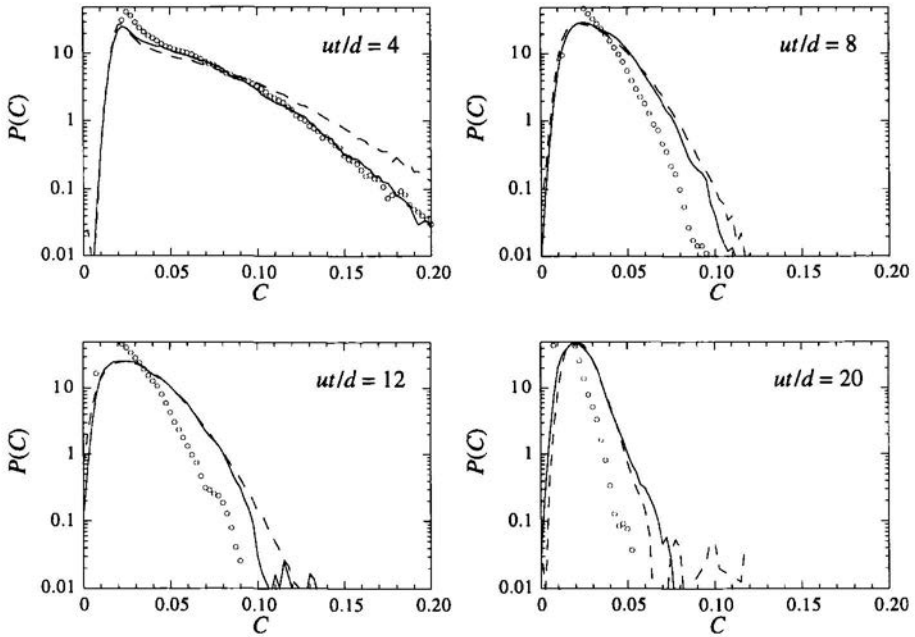


Figure 8 Composition of sources of heat in water ($Sc = 7$). Solid line: $P_{1+2}(C)$, experiment with two sources. Dashed line: reconstruction of the scalar field from $P_1(C)$ and $P_2(C)$ under the anticorrelated rule. Circles: reconstruction of the scalar field by convolution.

This result is extended to the turbulent jet problem (Duplat and Villiermaux 2000), for which it is shown that the scalar fluctuation PDF on the centerline is the result of well defined elementary sources whose size is given by the local width of the jet.

These findings, bridging the kinetics and statistical aspects of the phenomenon of mixing, also suggest that a complex mixture can be understood as the superposition of well defined elementary contributions, whose spatial distribution reflects the entrainment properties of the flow.

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