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Mixing versus Stirring

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Abstract

Mixing is the operation by which a system evolves under stirring from one state of simplicity –the initial segregation of the constituents–, to another state of simplicity –their complete uniformity–. Between these extremes, patterns emerge, possibly interact, and die sooner or later. This review summarizes the recent developments on the problem of mixing in its lamellar representation. This point of view visualizes a mixture as a set of stretched lamella, or sheets, possibly interacting with each other. It relies on a near-exact formulation of the Fourier equation on a moving substrate and allows to bridge the spatial structure and evolution of the concentration field with its statistical content in a direct way. Within this frame, both the dynamics of the concentration levels in a mixture as a function of the intensity of the stirring motions at the scale of a single lamella, and the interaction rule between adjacent lamella, are described precisely, thus offering a detailed representation of the mixture content, of its structure, and of their evolution in time.

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1. MIXING IS NOT BLENDING, NOR STIRRING

MÉLANGE: Il se dit de l'aggrégation de plusieurs choses diverses. *Encyclopédie*, Diderot & D'Alembert, 1765.

Mixing is the science describing the evolution of the concentration content in various substances (tracers, chemicals, heat, bacteria...) of a deforming continuum substrate. The subject matter is all contained in **Figure 1a** showing an initially concentrated blob of dye progressively incorporated in its diluting environment as the medium is stirred, down to a point where none of the constituents of the blob, and of the diluting phase, can be distinguished; they are mixed.

Mixing is the operation by which a system evolves from one state of simplicity –the initial segregation of the constituents–, to another state of simplicity –their complete uniformity–. Between these two extremes, patterns emerge depending on how the medium is deformed, possibly interact depending on how the mixture disperses in space, and die sooner or later depending on how Brownian noise has blurred the patterns on the way. As such, mixing is a paradigm of irreversible phenomena (Gibbs 1901).

1.1. Concentrations, inter-molecular scale, and fluctuations

Mixing deals with concentration fields, not with discrete particles. We first discuss the relationship between the spatial density of particles diffusing on a substrate, and the associated lengthscales when the substrate is stirred, to ensure the validity of a continuum description.

1.1.1. Concentrations. From a set of discrete particles sparsely spread in space with inter-particles distance λ , a number density $1/\lambda^3$ can be defined from their (molar) concentration c as

$$c = \frac{1}{\mathcal{N}\lambda^3} \quad 1.$$



Figure 1

a) A blob of ink deposited in glycerol is stirred by the sequential passage of a rod as a straw in a milk-shake would do. The formation of stretched lamella, getting thinner and overlapping as concentration differences fade away through the stirring cycles, is obvious (Villermaux & Duplat 2003). b) This celebrated picture from the very influential book by Arnold & Avez (1967) illustrates *blending*, not *mixing*, although the authors used the word ‘Mixing’ to refer to iterated maps distorting/spreading a blob in pieces with a probability of presence uniform in space. c) A typical random mixture in 2-d. A solitary strip unevenly stretched presents broad concentration fluctuations, and overlaps with itself in some places of the bounded stirring area.

where $\mathcal{N} \simeq 6.02 \times 10^{23}$ is the Avogadro number. For, say, molecules of a chemical species diluted in a liquid with concentration $c = 10^{-1} \text{ mol. l}^{-1}$, we have $\lambda \approx 10^{-8} \text{ m}$, a distance 10 to 100 times larger than the typical size of the molecules. A well defined c thus requires an averaging volume with size η substantially larger than λ . There are, in the mean, $\langle n \rangle \sim (\eta/\lambda)^3$ molecules randomly placed in this volume, with a relative number Poisson fluctuation of order

$$\frac{\sqrt{\langle n^2 \rangle - \langle n \rangle^2}}{\langle n \rangle} \sim \frac{1}{\sqrt{\langle n \rangle}}. \quad 2.$$

A representative concentration c defining a continuum exempt from trivial particle number fluctuations thus requires that $\eta \gg \lambda$.

In continuous media, discrete particles suffer Brownian agitation giving rise to the phenomenon of diffusion, whose intensity is measured by the diffusion coefficient D . For in-

stance, $D \sim (k_B T)^{3/2} / (a^2 p \sqrt{m})$ in a gas of molecules with size a and mass m at pressure p and temperature T , where k_B is the Boltzmann constant, or $D = k_B T / (6\pi\mu a)$ in a liquid with shear viscosity μ (Landau & Lifshitz 1987). When released around a point in a medium at rest, a set of particles will spread in time t over an isotropic cloud of radius \sqrt{Dt} , and if the medium is deformed (elongated and compressed) at a rate γ , we will show in section 2.4 that spreading is arrested in the compressive direction when the cloud has reached a transverse size

$$\eta = \sqrt{D/\gamma} \quad 3.$$

called the Batchelor (1959) scale. The condition for a smooth, well defined concentration field in a mixture is thus that the diffusion length $\sqrt{D/\gamma}$ (we will see in section 4.3.1 that this occurs at an even larger scale in complex mixtures) is larger than the inter-molecular distance of the species being mixed or, in other words, that the Péclet number

$$Pe_\lambda = \frac{\gamma \lambda^2}{D} \quad 4.$$

is smaller than unity. In liquids where the diffusion of big molecules is slow ($D \sim 10^{-9} \text{ m}^2 \text{ s}^{-1}$ or less), the Batchelor scale may be as small as a micron (10^{-6} m) while still remaining large enough to fulfill the condition of a continuum. The concentration, or scalar field c is then ruled by the conservation equation

$$\partial_t c + \nabla \cdot (\mathbf{v} c) = D \nabla^2 c \quad 5.$$

under the action of the stirring velocity field \mathbf{v} which may not be divergence free (i.e. $\nabla \cdot \mathbf{v} = 0$), and may or may not depend on c itself. The latter case is termed passive scalar mixing. The word ‘scalar’, as opposed to ‘vector’ (although very similar results as those discussed here exist for the magnetic field in 2-d flows, see Moffatt (1983); Childress & Gilbert (1995)), was employed, presumably for the first time in this context, by L. Kovasznay in 1961 at the Marseille symposium on the ‘Mechanics of Turbulence’ (Favre 1962).

1.1.2. The need for distributions of concentration. Examples abound showing that in most instances involving a mixing operation, it is not the mean concentration of the mixture $\langle c \rangle$ which is of interest, not even the standard deviation $\sqrt{\langle (c - \langle c \rangle)^2 \rangle}$ about the mean (Danckwerts 1952), but the probability of an extreme concentration event: The size of a combustion chamber, or of a chemical reactor will be set by the residence time of the mixture for the strongest, according to a desired criterium, concentration fluctuation to be erased (Marble 1964). Inhabitants living close to a leaking nuclear or chemical plant care if the concentration in pollutants of the effluents released by the leak, in air or through the ground (Csanady 1973), will be above or below the lethal dose when the pollutant plume reaches them, even if once in a lifetime. A remaining imperfection of additives composition in a glass, or a cement will be the weak link spoiling e.g. their mechanical resistance (Vidick 1989). Even the lifetime of liquid films as in sea bubbles is presumed to be set by highly concentrated impurities occurring with low probability in the liquid (Poulain *et al.* 2018).

Conversely, it is sometimes fortunate that a substance, or a blend transported by a flow has not mixed yet as patchy, intermittent concentrated regions might be vital clues: A bacteria (Berg 2004), a moth (Mafra-Neto & Cardé 1994) or a lobster (Koehl *et al.* 2001) directs its motion towards the source of pheromone or nutrients by sensing their

concentration above a detection level only (Schnitzer *et al.* 1990), which may be way above the mean concentration in the medium (Celani *et al.* 2014). The chemical composition of the rocks in the lithosphere, homogeneous along stripes but segregated from the rest of the mantle because yet unmixed offers precious clues about the early interior Earth (Allègre & Turcotte 1986).

This pressing reality, in conjunction with the fact that in science a satisfactory theory of a physical phenomenon requires its statistical description (Shraiman & Siggia (2000), think of the kinetic theory of gases (Maxwell 1867), or of Brownian motion (Chandrasekhar 1943; Reif 1965), for instance), leads us to focus on the distribution of concentration of the mixture $p(c)$, also called Probability Density Function, or PDF, such that

$$p(c)dc \tag{6}$$

is the probability to find in the mixture a concentration level between c and $c+dc$; obviously, $\int p(c)dc = 1$. The goal is to understand the construction mechanisms of $p(c)$ and to relate them to the detailed microscopic processes occurring in the mixture. The distribution $p(\Delta c)$ of the concentration increments $\Delta c(\Delta \mathbf{x}) = \langle c(\mathbf{x} + \Delta \mathbf{x}) - c(\mathbf{x}) \rangle$ is a quantity of general interest which presents original scaling properties (Kraichnan 1994; Falkovich *et al.* 2001) that we will consider too.

1.2. Semantics, misconceptions, and the singular role of diffusion

The subject matter is associated to a number of preconceptions and mental images that we consider now.

1.2.1. Semantics. An object may be defined by its opposite, and before we proceed to explain what mixing actually is, let us explain what it is not :

- Mixing is *not blending*, although a mixture is likely to mix well if it has been homogeneously blended. A perfectly well blended mixture might not be mixed at all if the constituents remain segregated from each other, even within finely divided domains. Mixing requires concentration homogeneity at the molecular scale λ . Imagine for example a mixture of particles with zero diffusivity ($D = 0$, an illusory limit in nature) which has been prepared in such a way that regions marked with $c = 1$ are adjacent to regions with $c = 0$. The marked regions are in relative proportion $\langle c \rangle$. The concentration distribution of the mixture is

$$p(c) = (1 - \langle c \rangle) \delta(c - 0) + \langle c \rangle \delta(c - 1) \tag{7}$$

and remains unchanged whatever the spatial reorganization of the field and the division state of the mixture may be; indeed, whatever \mathbf{v} may be provided it is incompressible (i. e. $\nabla \cdot \mathbf{v} = 0$, no net expansion nor contraction of the substrate), c is ruled by $\partial_t c + \mathbf{v} \cdot \nabla c = 0$ and is conserved along Lagrangian trajectories. There is, in this instance, no mixing at all. There would be if $D \neq 0$ and in that case sustained motions of the substrate would ultimately lead to

$$p(c) \xrightarrow[t \rightarrow \infty]{} \delta(c - \langle c \rangle) \tag{8}$$

or approach this perfectly well mixed limit after a *mixing time* which deserves to be understood in terms of the nature of the stirring motions.

- Mixing is *not stirring*, although a vigorously stirred mixture will reach homogeneity faster than a mixture kept at rest, this one being sensitive to the typically slow (these adjectives will be quantified later) molecular diffusion only. Stirring may contribute to efficient blending as in **Figure 1b** taken from Arnold & Avez (1967), but stirring alone will not, for the reason underlined above, mix (see the lucid statements in Brodkey (1967), and Epstein (1990)).

1.2.2. The singular role of diffusion. Mixing is stretching enhanced diffusion: Concentration change ($\partial_t c$) results from a subtle coupling between advection ($\mathbf{v} \cdot \nabla c$) and diffusion ($D\nabla^2 c$) in equation (5.), and fluctuations about the mean will decay according to (Zeldovich 1937)

$$\frac{d}{dt} (\langle c^2 \rangle - \langle c \rangle^2) = -2D\langle (\nabla c)^2 \rangle \quad 9.$$

only if $D \neq 0$. This singular role of molecular diffusion, and its coupling with substrate motions is familiar, particularly in the context of dispersion. It is known that without diffusion, the second moment of the residence time distribution of a tracer dispersing along a laminar pipe (radius h , mean velocity U), diverges. It is finite, with an effective longitudinal dispersion coefficient $D_{\text{eff}} \sim DPe^2$ with $Pe = Uh/D$ as soon as $Pe < \infty$ (Taylor 1953). In cellular flows, like along an array of stationary convection cells, the only way a dye can jump from one cell to the other is by crossing their separatrices by molecular diffusion, and in that case $D_{\text{eff}} \sim D\sqrt{Pe}$ (Shraiman 1987; Solomon & Gollub 1988; Biferale *et al.* 1995), a conclusion which holds also for reactive mixtures (Audoly *et al.* 2000). Periodic oscillations of the separatrix location allow for cells to exchange material according to a mechanism imagined by Melnikov in 1963 (Rom-Kedar *et al.* 1990) but this ‘lobe dynamics’ will not alter, alone, the mixture composition (Beigie *et al.* 1991). In layered systems, like porous rocks with broad permeability distributions, it is the diffusion across the layers which regularizes the dispersion process along the layers, which otherwise would be purely ballistic (Matheron & de Marsilly 1980; Bouchaud & Georges 1990).

The existence of diffusion, even by a tiny amount, changes paradigm.

1.2.3. History matters. If mixing is contingent upon diffusion, a frequent underlying assumption, not firmly formalized as such (Sturman *et al.* 2006; Aref & al. 2017), is that since stirring and diffusion are in essence two different phenomena (which is true), it is therefore paramount to focus on how material particles are advected, because a simili-diffusion can always be incorporated in the end of the advection process, by some local coarsening, to account for the ‘smearing’ of the –yet unmixed– scalar field. This sequential vision is fundamentally incorrect, as we show on hand of the following example (sidebar below).

The whole stretching history, inherently coupled with the permanent, but possibly enhanced, or slowed down, action of diffusion has to be accounted for in a precise representation a mixture’s fate.

1.3. Approach and scope

This review summarizes the recent developments on the problem of mixing in its lamellar representation (Batchelor 1959; Ranz 1979; Ottino 1982). This point of view, which visualizes a mixture as a set of stretched lamella, or sheets, possibly interacting with each other is extremely powerful since it relies on a near-exact formulation of the Fourier equation on a moving substrate (5.), and because it allows to bridge the spatial structure and evolution of

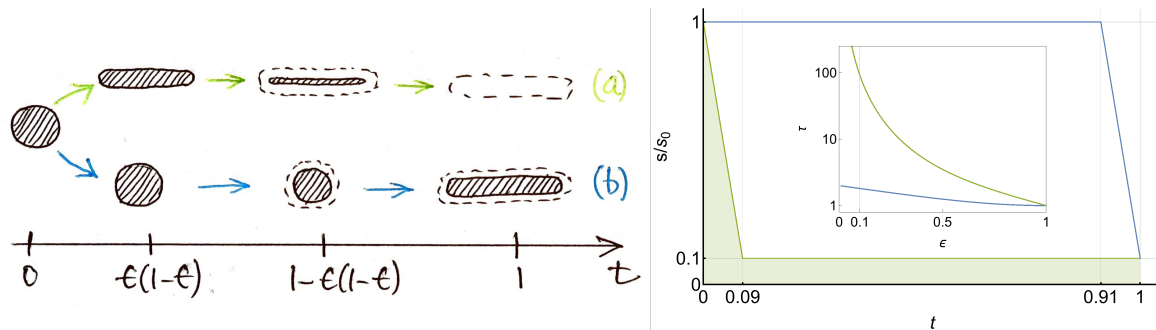


Figure 2

History matters: The two kinematically identical deformation protocols of a diffusing blob, and their very different mixing states in $t = 1$ (see text).

HISTORY MATTERS: AN EXAMPLE

A blob of size s_0 is deposited at $t = 0$ on a 2-d substrate. At the same time the blob diffuses, we stretch the medium so that the blob elongates in one direction, and compresses in the other down to, say, $1/10^{\text{th}}$ of its initial size. We do this according to two distinct protocols. In (a), we first squeeze the blob within a short time according to $s(t) = s_0(1 - t/\epsilon)$ down to $s(t_\epsilon)/s_0 = \epsilon$, and then let it still up to $t = 1$ (in units of the blob pure diffusion time s_0^2/D). In (b), we let the blob still during a time $1 - t_\epsilon$, and then squeeze it in the same way during $t_\epsilon = \epsilon(1 - \epsilon)$. Since $\epsilon \ll 1$, the squeezing motion is at large Péclet number $Pe = \epsilon^{-1}$. The deformation kinematics of the blobs are identical in both protocols and in the absence of diffusion, (a) and (b) would be undistinguishable. But diffusion has operated on the way and it is not difficult to anticipate that protocol (a) is more efficient at decaying the concentration in the stretched blob than (b), for which the blob has remained thick for most of the time. We will see in section 2.3 that the maximal concentration of the blob is given by $\text{erf}(1/4\sqrt{\tau})$ with $\tau \sim \int_0^1 dt'/s(t')^2$, and **Figure 2** demonstrates that in protocol (a), $\tau \sim \epsilon^{-2}$ is 50 times larger than in (b) where $\tau \sim 2$. The blob concentration is therefore 2.6 times larger in (b) than in (a) at $t = 1$ for $\epsilon = 0.1$, a ratio which is even larger for a smaller ϵ . History matters.

the concentration field with its statistical content in an direct way (Meunier & Villermaux 2003). Within this frame, both the dynamics of the concentration levels in a mixture as a function of the intensity of the stirring motions at the scale of a single lamella, and the nature of the interaction rule between adjacent lamella, are described precisely (Villermaux & Duplat 2003). This offers a detailed description of the mixture concentration content $p(c)$ (Duplat & Villermaux 2008), of its structure $p(\Delta c)$ (Le Borgne *et al.* 2017), and of their evolution in time.

2. STRETCHING ENHANCED DIFFUSION

Because the displacement gradients of the stirring motion typically form elongated structures (lamella in 2-d, sheets in 3-d) from an initially compact isotropic blob passively advected by the flow, concentration gradients are, usually, only notable in the direction

perpendicular to the direction of elongation (at the exception of rare highly curved regions of the scalar support (Thiffeault 2004), which are ever rarer as stirring proceeds (Meunier & Villermaux 2010)). This is the reason why a one-dimensional description of the local concentration field dynamics is relevant, as it captures the essence of the coupling between stretching rate, and scalar decay.

2.1. Diffusion on still, and moving substrates

We first recall when and how stirring the substrate affects diffusion.

2.1.1. Still substrate. The concentration $c(x, t)$ at position x and time t of N diffusing particles released at the origin of an axis in $t = 0$, i.e. $c(x, 0) = N\delta(x)$ is (Fourier 1822)

$$c(x, t) = \frac{N}{2\sqrt{\pi Dt}} e^{-\frac{x^2}{4Dt}} \quad 10.$$

while if $N = c_0 s_0$ particles have been deposited with uniform concentration c_0 in the interval $x \in \{-s_0/2, s_0/2\}$ at $t = 0$ like for a blob of width s_0 , one has by integration of the Green's function $c(x - x', t)$ in (10.) on the interval (see e.g. Carslaw & Jaeger (1986))

$$c(x, t) = \frac{c_0}{2} \left\{ \operatorname{erf} \left(\frac{x + s_0/2}{\sqrt{4Dt}} \right) - \operatorname{erf} \left(\frac{x - s_0/2}{\sqrt{4Dt}} \right) \right\} \quad 11.$$

The long time limit $\sqrt{Dt} \gg s_0$ of (11.) recovers (10.). The maximal concentration $c(0, t) \equiv \theta(t)$ at the center of the blob in $x = 0$ is

$$\theta(t) = c_0 \operatorname{erf} \left(\frac{s_0/2}{\sqrt{4Dt}} \right) \quad 12.$$

$$\sim \frac{c_0 s_0}{\sqrt{Dt}}, \quad \text{for } t \gg t_s \simeq \frac{s_0^2}{D} \quad 13.$$

an asymptotic trend expressing mass conservation which generalizes to d-dimensions as $\theta(t) \sim c_0 \left(s_0 / \sqrt{Dt} \right)^d$, holding after the d-independent *mixing time* $t_s \simeq s_0^2/D$ beyond which the concentration in the blob has departed appreciably from its initial value to reach the asymptotic decay.

The discussion in the sequel is not affected by the particular choice we have made for the initial condition in (11.) which is only meant to isolate a blob with uniform concentration from its diluting environment where $c = 0$. A blob defined by a Gaussian concentration profile of width s_0 (Meunier & Villermaux 2010), or any other shape leads to identical considerations.

2.1.2. Moving substrate. On a stirred substrate, diffusion competes with the deformation of the medium. The diffusion flux $-D\nabla c$ is proportional to the concentration gradient that is, essentially, to the concentration difference between two points. If these points get further apart like in the stretching directions of the substrate, the gradient decays, and so does the flux. In compressive regions, the gradient steepens, and the diffusion flux is enhanced. For this key mechanism to operate, substrate compression must be fast enough.

Any kind of shear motion will elongate a blob into a lamella, say of width of order s . The lamella will decay by transverse diffusion according to equation (10.) in a time s^2/D . If

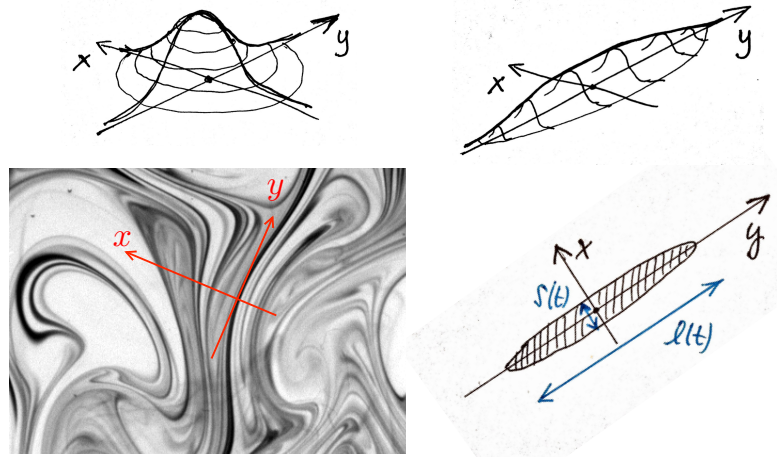


Figure 3

Diffusion on a moving substrate. A blob which would otherwise expand isotropically is stretched along y , and compressed along x , the permanent process forming lamella in stirred mixtures.

at the same time the substrate is compressed at a rate γ such that $\dot{s} = -\gamma s$, it is clear that γ^{-1} should be smaller than s^2/D for gradient reinforcement to be effective. Thus, starting with a blob of size s_0 , mixing will amount to a simple diffusion problem if $s_0^2/D \ll \gamma^{-1}$, and a genuine non-trivial coupling will occur when $s_0^2/D \gg \gamma^{-1}$, that is when the Péclet number

$$Pe = \frac{\gamma s_0^2}{D} \quad 14.$$

is larger than unity.

2.2. The Ranz transformation

In a local Lagrangian frame $\{x, y\}$ moving with a lamella (in 2-d, the discussion is readily generalized to a sheet in 3-d (Martinez-Ruiz *et al.* 2018)) so that the direction x points in the direction of the maximal concentration gradient, and y is perpendicular to it (these directions tend to align with the eigenvectors of the deformation tensor, see Ashurst *et al.* (1987)), the components $\{u, v\}$ of the velocity field \mathbf{v} in equation (5.) are related to the compression rate of the (incompressible, $\nabla \cdot \mathbf{v} = 0$) substrate material particles by $u = (\dot{s}/s)x$ and $v = -(\dot{s}/s)y$.

Given the discussion above on the relative magnitude of the concentration gradients in the elongating, and compressing directions of the substrate, we see that the local dynamics of the concentration field from equation (5.)

$$\partial_t c + u \partial_x c + v \partial_y c = D (\partial_x^2 + \partial_y^2) c \quad 15.$$

incorporates two sub-dominant terms. Under stretch at large Pe , a blob of initial surface s_0^2 is elongated into a strip of width s and length $\ell \gg s$ (see **Figure 3**) so that, by incompressibility $s_0^2 \sim s \times \ell$. The orders of magnitude of the components of the concentration gradient are $\mathcal{O}(\partial_x c) = 1/s$ and $\mathcal{O}(\partial_y c) = 1/\ell$. Thus, in a neighborhood of size s at the

Stirring protocols and mixing times

We list below some standard stirring protocols, along with their mixing time t_s from the condition (21.), and the maximal concentration $\theta(t)$ in (20.) for $t > t_s$, in the large Pe limit.

For a simple shear (Ranz 1979), we have $s(t) = s_0/\sqrt{1 + (\gamma t)^2}$, providing

$$\tau = \frac{\gamma t}{Pe} \left(1 + \frac{1}{3}(\gamma t)^2 \right), \quad \text{giving} \quad t_s \sim \frac{1}{\gamma} Pe^{1/3}, \quad \text{and} \quad \theta(t) \sim (\gamma t)^{-3/2}.$$

Elongations in two directions (Okubo & Karweit 1969) with $s(t) = s_0/(1 + (\gamma t)^2)$ provide

$$\tau = \frac{\gamma t}{Pe} \left(1 + \frac{2}{3}(\gamma t)^2 + \frac{1}{5}(\gamma t)^4 \right), \quad \text{giving} \quad t_s \sim \frac{1}{\gamma} Pe^{1/5}, \quad \text{and} \quad \theta(t) \sim (\gamma t)^{-5/2}.$$

A stagnation (saddle) point with steady stretching (Batchelor 1959) is such that $s(t) = s_0 e^{-\gamma t}$, thus

$$\tau = \frac{1}{2Pe} (e^{2\gamma t} - 1), \quad \text{giving} \quad t_s = \frac{1}{2\gamma} \ln(1 + 2Pe) \xrightarrow{Pe \gg 1} \frac{1}{2\gamma} \ln Pe, \quad \text{and} \quad \theta(t) \sim e^{-\gamma t},$$

where it should be noted that the pure diffusive limit $t_s \sim s_0^2/D$ is recovered for $Pe \ll 1$ and that, under stretch, diffusion is arrested in the elongating direction where $\ell(t) = s_0 e^{\gamma t}$ since in that case $\tau = \frac{1}{2Pe}(1 - e^{-2\gamma t}) \rightarrow 1/2Pe$.

Sub ($\alpha < 1$) or super ($\alpha > 1$) exponential stretching (de Rivas & Villiermaux 2016) with $s(t) = s_0 e^{-(\gamma t)^\alpha}$ correspond to

$$\tau \sim \frac{1}{Pe} \frac{e^{2(\gamma t)^\alpha}}{2\alpha(\gamma t)^\alpha}, \quad \text{giving} \quad t_s \sim \frac{1}{\gamma} (\ln Pe)^{1/\alpha}, \quad \text{and} \quad \theta(t) \sim e^{-(\gamma t)^\alpha}.$$

A finite time singularity (Villiermaux 2012b) such that $s(t) = s_0(1 - \gamma t)^\alpha$ with $\alpha > 1/2$ gives

$$\tau = \frac{1 - (1 - \gamma t)^{1-2\alpha}}{(1 - 2\alpha)Pe}, \quad \text{giving} \quad \gamma t_s \sim 1 - Pe^{1/(1-2\alpha)} \xrightarrow{Pe \rightarrow \infty, \alpha > 1/2} 1.$$

It is the only instance where the mixing time t_s remains *strictly* finite at $Pe = \infty$, and given by the singularity time γ^{-1} at which $s(\gamma^{-1}) = \theta(\gamma^{-1}) = 0$.

center of the strip $\mathcal{O}(|u\partial_x c|/|v\partial_y c|) = \ell/s \gg 1$ and for the same reason, $\mathcal{O}(|\partial_x^2 c|/|\partial_y^2 c|) = (\ell/s)^2 \gg 1$. The *near exact* (at large Pe) form of the evolution equation for c is thus

$$\partial_t c + (\dot{s}/s) x \partial_x c = D \partial_x^2 c \tag{16.}$$

As it is, (16.) already represents a considerable progress since it bridges, by a linear equation, the dynamics of c with the prescribed kinematics of the stirring field through a single feature, namely the compression rate \dot{s}/s . This compression rate reflects, in incompressible flows, the growth rate of material lines length (i.e. $\dot{\ell}/\ell$), or the area of surfaces.

More can be done towards a deeper understanding of (16.), a step which also makes it more practical to use. Distances x and time t define the space in which we discuss physical phenomena, but it is not necessarily the natural one. A coordinate change, popularized by Marble & Broadwell (1977) and Ranz (1979) consisting in counting distances in units of s , and time in units of the diffusion time s^2/D as

$$\xi = \frac{x}{s(t)}, \quad \text{and} \quad \tau = D \int_0^t \frac{dt'}{s(t')^2} \quad 17.$$

transforms equation (16.) into a pure diffusion equation

$$\partial_\tau c = \partial_\xi^2 c \quad 18.$$

This extremely elegant, and useful result is consistent with the fact that only molecular diffusion can alter the concentration content of a field; it is thus natural that *in fine*, the dynamics of c complies to pure diffusion, the dilatation, or compression of space (i.e. the time dependence of $s(t)$) being just a way to delay, or hasten the process. Since $s(t)$ typically decreases in time under stirring, τ in (17.) increases faster than linearly in time, expressing the expected acceleration of diffusion.

Either in its original, or in slightly different forms, equation (18.) found its use in various disciplines ranging from heat transfer (Levêque 1928), turbulence (Batchelor 1959), reacting flows (Gibson & Libby (1972); Carrier *et al.* (1975); Marble (1988)), engineering and process industry (Mohr *et al.* 1957; Ranz 1979), geophysics (Rhines & Young 1983; Allègre & Turcotte 1986), chaos (Ottino 1982; Beigie *et al.* 1991), physics (Moffatt 1983), or mathematics (Fannjiang *et al.* 2004).

From a blob, or strip of initial transverse size s_0 , the concentration (scaled by c_0) in the genuine coordinates (17.) is

$$c(\xi, \tau) = \frac{1}{2} \left\{ \operatorname{erf} \left(\frac{\xi + 1/2}{2\sqrt{\tau}} \right) - \operatorname{erf} \left(\frac{\xi - 1/2}{2\sqrt{\tau}} \right) \right\} \quad 19.$$

obtained in the same way the concentration profile in (11.) was.

2.3. Maximal concentration and mixing time

The maximal concentration in the lamella is found in $x = 0$, that is $\xi = 0$ and is

$$\theta(\tau) = \operatorname{erf} \left(\frac{1}{4\sqrt{\tau}} \right) \xrightarrow{\tau \gg 1} \frac{1}{\sqrt{\tau}} \quad 20.$$

while $\theta(\tau) \approx 1$ as long as $\tau \ll 1$. In line with section 2.1.1, we define the *mixing time* t_s beyond which the concentration in the lamella has departed appreciably from its initial value to reach the asymptotic decay $1/\sqrt{\tau}$ by the condition

$$\tau(t_s) = \mathcal{O}(1) \quad 21.$$

To any stirring protocol, involving any particular form of $s(t)$, and therefore of $\tau(t)$, corresponds a given t_s with, notably, a given dependence on Pe (see the sidebar 2.2). The mixing time is always of the form

$$t_s \sim \frac{1}{\gamma} \mathcal{F}(Pe) \quad 22.$$

where γ relates to the deformation rate of the substrate, and $\mathcal{F}(Pe)$ is a weak function of the Péclet number Pe , typically a small power, or a logarithm, a fact known to engineers for a long time (Nagata 1975). Irrespective of the nature of the substance being mixed (i.e. of D), the time it takes to mix it in a stirred vessel with a standard impeller rotating \dot{N} rounds per unit of time is $\dot{N}t_s \approx 5$, the factor 5 reflecting geometrical factors, and a logarithmic Péclet correction at large Reynolds number which is so weak that it is insensitive in the engineering practice.

Said differently in order to allude to an image frequently associated with mixing, it is well known that when stirring a drop of milk in a cup of coffee, only the number of spoon turns matters. This fact is indeed familiar in the turbulence context (at large Reynolds number, the analogue of the Péclet number for vorticity (Moffatt 1983)) where the ‘cascade time’ from the blob injection (of scalar, or vorticity) to its dissipation by molecular diffusion, or viscosity is essentially independent of them, or involves a weak correction only.

For being weak in practice since a logarithm is close to a constant at large Pe (see e.g. Donzis *et al.* (2005)), the correction $\mathcal{F}(Pe)$ is nevertheless singular (it is infinite in the limit $D \rightarrow 0$, sometimes coined a ‘dissipative anomaly’ (Falkovich *et al.* 2001)), and this makes sense : The decay *rate* of the scalar fluctuations is solely prescribed by the stirring strength γ , but it is so *after* the mixing time only. In other words, it takes some time for the stretching motions to bring the scalar blob down to a scale small enough for molecular diffusion to become effective in erasing the scalar differences. This time depends both on the stirring strength γ , on the initial blob size s_0 , *and* on its diffusional properties D ; this is the essence of mixing in stirred media.

After the mixing time, the maximal concentration $\theta(t)$ in (20.) decreases according to mass conservation $\theta(t)\sqrt{Dt}/s(t) \sim 1$ as $\theta(t) \sim (\gamma t)^{-\alpha-1/2}$ for power law stretching, or $\theta(t) \sim e^{-\gamma t}$ with exponential stretching (see the sidebar 2.2), and in any case faster than for pure diffusion where $\theta(t) \sim (Dt)^{-d/2}$ in d-dimensions.

2.3.1. The case of small Péclet numbers. We have stressed in section 2.1.2 that mixing is a non-trivial problem in the limit of large Péclet numbers only, but this is not exactly true. An interesting coupling occurs for $Pe < 1$ in a shear flow: At low Pe , diffusion broadens a (small) blob in its traverse direction as $\sqrt{Dt} > s_0$ resulting, since the blob sits in a shear, in a longitudinal dispersion velocity of the blob $\dot{\ell} \sim \gamma\sqrt{Dt}$, that is a blob length scaling like $\ell(t) \sim \gamma\sqrt{D}t^{3/2}$ (this is besides a well-known recipe to produce a super-diffusive dispersion law *à la* Richardson (1926), see for instance Celani *et al.* (2005)). The blob surface increases like $\sqrt{Dt} \times \ell(t) = \gamma Dt^2$ which, by mass conservation, provides the maximal concentration carried by the blob $\theta(t)$ as

$$\theta(t) \sim \frac{s_0^2}{\gamma Dt^2} \tag{23.}$$

This régime is however likely to operate if $\ell(t)$ above is larger than the pure kinematic elongation of the blob $s_0\gamma t$, that is for $t > s_0^2/D$, a time large compared to $t_s \sim Pe^{1/3}/\gamma$ unless $Pe < 1$. The corresponding régime will thus affect the early dynamics of a blob for Pe , at best, of order unity. A rigorous treatment of this nice exercise can be found in (Thiffeault 2008).

Rule of Thumb

The qualitative meaning of the crossover condition in equation (21.) is that diffusion starts to operate when its associated timescale compares to the substrate deformation time, that is

$$|\dot{s}/s| \sim D/s^2$$

Taking generically $s(t) \sim s_0(\gamma t)^{-\alpha}$, one sees that

$$t_s \sim \frac{1}{\gamma} Pe^{1/(2\alpha+1)}, \quad \text{and} \quad \theta(t) \sim (\gamma t)^{-\alpha-1/2}$$

explaining why the Pe -dependence of t_s is weaker when the stretching is stronger (i.e. large α).

2.4. Batchelor scales and dissipation

We have emphasized how diffusion broadening competes with substrate compression. To this process is associated a lengthscale η , which we have already alluded to in section 1.1.1. There is in fact a family of lengthscales, all representative of the same phenomenon. For $\tau > 1$, that is for $t > t_s$, the concentration profile across a lamella (19.) converges towards a decaying Gaussian (de Rivas & Villiermaux 2016)

$$c(x, t) \sim \frac{1}{2\sqrt{\pi\tau}} e^{-\frac{x^2}{2\eta^2}} \quad 24.$$

$$\text{with } \eta(t) = s(t)\sqrt{\tau(t)} \quad 25.$$

We call $\eta(t)$ and, more precisely $\eta(t_s)$, a Batchelor scale. After the mixing time, $\eta(t)$ has no reason to be a constant, in general. It is, in the special stirring protocol of a constant stretching rate γ . In that case, $s(t) = s_0 e^{-\gamma t}$ and $\tau(t) \sim e^{2\gamma t}/Pe$, therefore $s(t)\sqrt{\tau(t)} \sim \sqrt{D}/\gamma$ is indeed a constant of time, also independent of s_0 (Batchelor 1959).

This lengthscale first arose in turbulence, where the relevant stretching rate $\gamma = (U/L)Re^{1/2}$ is the one prevailing for scales below the Kolmogorov scale $LRe^{-3/4}$ where $Re = UL/\nu$ is the Reynolds number with U a velocity at the large scale L and ν the fluid kinematic viscosity. The (original) Batchelor scale $LRe^{-3/4}Sc^{-1/2}$ with $Sc = \nu/D$ the Schmidt number is usually difficult to detect in flows at large Reynolds, and Schmidt numbers (note that $Pe = Re \times Sc$), because it is small (Miller & Dimotakis 1996); it is however more easily accessible to precise numerical simulations (Schumacher *et al.* 2005), or to experiments involving moderate, and simple deformations fields (Meunier *et al.* 2015). Anticipating on section 3.2, we note that since in random flows stretching rates are distributed in intensity, the Batchelor scale above has to be understood as a representative mean of an otherwise broad distribution of scalar dissipation scales (Schumacher *et al.* 2005).

In more general stirring protocols, the compression rate $\gamma(t) = -\dot{s}/s$ is itself time-dependent. For instance, if $s(t)$ decreases as a power law like $s(t) \sim s_0(\gamma t)^{-\alpha}$, one has

$$\eta(t) \sim \sqrt{Dt} \quad 26.$$

consistent with the large time decay of the compression rate $\gamma(t) \sim 1/t$, finally overcome by diffusion broadening. Since in that case (see the rule of thumb in 2.3.1) $\gamma t_s \sim Pe^{1/(2\alpha+1)}$,

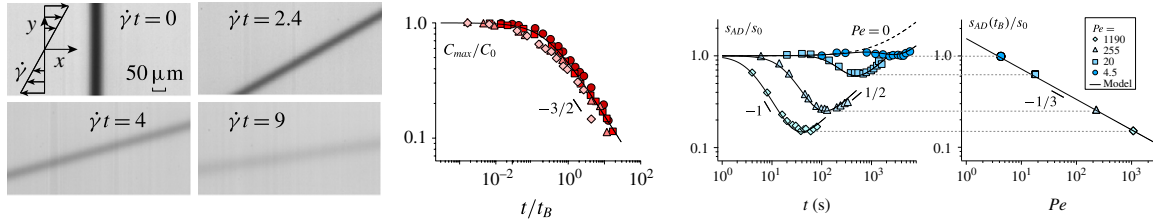


Figure 4

a) Lamella mixing in a shear flow. b) Decay of the maximal concentration $\theta(t) \sim (t/t_s)^{-3/2}$, c) Standard deviation of the lamella concentration profile exhibiting first kinematic compression, then diffusive broadening according to (26.) and d) Péclet number dependence of the Batchelor scale in (27.) with $\alpha = 1$. From Souzy *et al.* (2018).

then

$$\eta(t_s) \sim s_0 Pe^{-\alpha/(2\alpha+1)} \quad 27.$$

now depending on the value of α , and on the initial condition s_0 . The case $\alpha = 1$ (shear flow) was precisely investigated by Souzy *et al.* (2018) who indeed confirmed all the necessary trends and scaling laws (**Figure 4**).

In time-dependent flows, if α is a number reflecting the accelerated nature of the stretch intensity ($\alpha > 1$), or its slowing down ($\alpha < 1$), we have

$$\frac{\dot{\gamma}(t)}{\gamma} = \alpha (\gamma t)^{\alpha-1}, \quad 28.$$

a formulation which has no other fundamental justification than being easily adaptable to different flow configurations knowing that in nature, diverse behaviors exist concomitantly, and/or sequentially (McKenzie 1979). The corresponding Batchelor scale in (27.) tends when $t > t_s$ towards

$$\eta(t) \sim \sqrt{\frac{D}{\alpha\gamma}} (\gamma t)^{\frac{1-\alpha}{2}} \quad 29.$$

and coincides with the usual constant value $\sqrt{D/\gamma}$ in exponentially diverging flows with $\alpha = 1$. The concentration gradient goes on steepening as the maximal concentration decays in the accelerated regions of the flow (see an example with $\alpha = 2$ in Néel & Villermaux (2018)), and relaxes for slowed down stretching (de Rivas & Villermaux 2016).

2.4.1. Dissipation. Among the many global, lumped indices which have been defined to quantify a mixing state, or the ‘mixideness’ of a given protocol that is its ability to mix well, such as the intensity of segregation (variance of c about the mean, Danckwerts (1952)), the dilution index (entropy of $p(c)$, Kitanidis (1994)), the mix-norm (field coarsening, Mathew *et al.* (2005) see also Thiffeault (2012)), which are all ersatz of the concentration distribution $p(c)$ (see Le Borgne *et al.* (2015)), is the dissipation rate $\chi(t) = -2D\langle(\nabla c)^2\rangle$. It is the average squared concentration gradient which, when weighted by D , is the decay rate of the mean squared concentration differences about the mean (see (9.), and Zeldovich (1937)).

From the Batchelor scale $\eta(t)$, and the maximal concentration in a lamella $\theta(t)$, a typical concentration gradient is $\theta(t)/\eta(t)$, and for an isolated stretching blob at $t \gg t_s$

$$\chi(t) \sim \gamma \sqrt{Pe} (\gamma t)^{-\alpha-3/2} \quad (\text{power law stretching}) \quad 30.$$

$$\chi(t) \sim \gamma \sqrt{Pe} e^{-\gamma t} \quad (\text{exponential stretching}) \quad 31.$$

exhibiting, not surprisingly, a stronger time-dependence than for pure diffusion where $\chi(t) \sim D^{-d/2} t^{-1-d/2}$ in d -dimensions.

There is no dissipation as $D \rightarrow 0$, because $t_s \rightarrow \infty$ in that singular limit (see section 2.3 and Balmforth & Young (2003)). The above relations are readily generalized to non-decaying mixtures where blobs are periodically injected and stirred at steady state (Villermaux 2012*b*). This is a way to understand from elementary principles eddy diffusivities, or heat and mass transfer laws at sheared boundaries.

3. SOLITARY STRIPS

By solitary strips we mean lamella carrying concentration levels (20.) solely prescribed by their local stretching history according to (17.). In that case, the concentration distribution $p(c)$ simply reflects the relative cumulated elongation intensities along the strip at a given time. We examine several examples with either steady, or time-dependent stirring protocols.

3.1. Deterministic stirring

We call deterministic those stirring protocols which are either steady, or time-dependent but which all lead to a unique trajectory of the deformed blob for a given initial condition.

3.1.1. The concentration distribution of a Gaussian spatial profile. We aim at giving a representation of the distribution $p(c)$ of the concentration levels c along a lamella distorted by a flow. After the mixing time, these levels are carried by the local Gaussian spatial profile across the lamella in (27.) parametrized by its maximum θ , and width η ; the concentration levels span from 0 far from the lamella, to θ . Exploring, along the axis x across the lamella (see (27.)), the c levels over a x -range of the order of a few η , each are encountered with a relative frequency given by

$$g(c|\theta) \sim \frac{\eta^{-1}}{|dc/dx|_{x(c)}} \sim \frac{1}{c\sqrt{\ln(\theta/c)}} \quad 32.$$

with $x(c) \sim \eta\sqrt{\ln(\theta/c)}$. The characteristic \cup shape of this distribution is well known (Meunier & Villermaux 2003, 2007, 2010; Duplat *et al.* 2010*a*; Martinez-Ruiz *et al.* 2018; Souzy *et al.* 2018). The distribution (32.) is however not normalized because of its divergence in $c = 0$, reflecting the free choice for defining the support of the lamella, which can extend arbitrarily far from it in its diluting ocean, a divergence which is thus not physically meaningful. The distribution $g(c|\theta)$ has another divergence in $c = \theta$ which singles out the concentration maximum, and it is this divergence which carries the relevant information given that, in non-trivial flows, θ is itself distributed.

Unless explicitly taken into account when they give rise to an interesting phenomenon (Meunier & Villermaux 2007), it is usually fair to disregard the contributions of the low concentration levels from the spatial tail of the Gaussian profile (27.), and thus approximate (32.) as

$$g(c|\theta) \approx \delta(c - \theta) \quad 33.$$

which serves our purpose to discuss the large excursion shape of the distribution $p(c)$ of a strip along which the maxima θ are distributed.

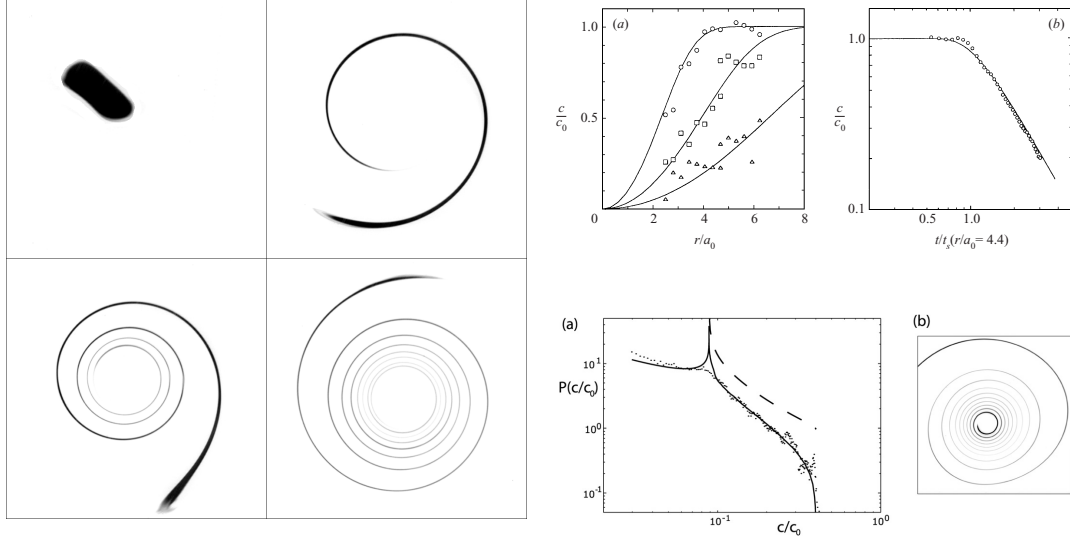


Figure 5

Mixing by a vortex. A) A blob in the far field of a Lamb-Oseen vortex spirals around the vortex. B) Concentration $\theta(r, t)$ at given instants of time along the spiral (a), and a fixed radial location versus time (b) together with the expected relation (continuous lines) from $\theta(r, t) = \text{erf}\left(1/\sqrt{4\tau(r, t)}\right)$ with $\tau(r, t)$ given in (35.). C) Concentration distribution of the spiraling blob at a given instant of time, and expected law (continuous line) presenting a Van Hove singularity when the blob is deposited close to the viscous core of the vortex. From Meunier & Villermaux (2003, 2007).

3.1.2. Mixing by a vortex. The stirring protocol of a permanent point vortex with circulation Γ (azimuthal velocity $\Gamma/2\pi r$) is an illustration of the construction mechanism of $p(c)$ which is worth considering in some details (Meunier & Villermaux 2003).

A blob of size s_0 is deposited at a distance $\tilde{r} \gg s_0$ from the center of the vortex. An element of surface $s_0 dr$ of the blob is stretched kinematically into a strip of length $d\ell$ such that

$$d\ell = dr \sqrt{1 + \frac{\Gamma^2 t^2}{\pi^2 r^4}} \rightarrow dr \frac{\Gamma t}{\pi r^2} \quad 34.$$

spiraling around the vortex center. Area conservation $s_0 dr = s(r, t) d\ell$ thus gives rise to a time-dependent, and radius dependent stretching rate, so that τ in (17.) depends on both r and t as

$$\tau(r, t) = \frac{Dt}{s_0^2} \left(1 + \frac{\Gamma^2 t^2}{3\pi^2 r^4}\right) \quad 35.$$

with a time-dependence (i.e. t^3) reminiscent of flows which increase material line lengths in proportion of time (the length of the spiral is $L = \int d\ell \approx \Gamma s_0 t / \tilde{r}^2$). The condition $\tau(r, t) = \mathcal{O}(1)$ provides a r -dependent mixing time

$$t_s(r) \sim \frac{r^2}{\Gamma} \left(\frac{s_0}{r}\right)^{2/3} \left(\frac{\Gamma}{D}\right)^{1/3}. \quad 36.$$

The fluid particles of the blob closer to the center of the vortex are stretched faster, and have hence a shorter mixing time; they also carry a smaller maximal concentration $\theta(r, t) = \text{erf}(1/4\sqrt{\tau}) \sim 1/\sqrt{\tau(r, t)}$ because they have mixed earlier (**Figure 5**). The one-to-one

correspondence between strip elongation, and maximal concentration along the deformed blob translates in the conservation law $q(\theta)d\theta = d\ell/L$, providing the distribution of maximal concentrations as

$$q(\theta) \sim \frac{\Gamma t}{Lr^2} \frac{1}{|d\theta/dr|} \quad 37.$$

The full concentration field c can be reconstructed precisely (Meunier & Villiermaux 2003) from the elementary \cup distributions in (32.), also describing some features like Van Hove singularities when the spatial concentration field presents a saddle point in a variant of the present problem (Meunier & Villiermaux 2007). At large times, the use of the approximation (33.) is such that $p(c) = \int g(c|\theta)q(\theta)d\theta \approx q(\theta = c)$, and when $t > t_s(r)$ for all $r \in \{\tilde{r}, \tilde{r} + s_0\}$

$$p(c) \sim \left(\frac{s_0}{\sqrt{D\Gamma t^{3/2}}} \right)^{1/2} \frac{\tilde{r}}{c^{3/2}} \quad 38.$$

Because the particles close to the vortex center are more stretched, they occupy a larger fraction of the spiral than the remote ones. Since their concentration is smaller as they are more elongated, then the overall $p(c)$ is a decreasing function of c . This simple fact is the paradigm of solitary strip mixing.

3.2. Random Flows

An identical construction as above, relating the strips elongations to $p(c)$ holds when stirring is time-dependent in the sense of chaotic or turbulent. Irrespective of the stirring protocol, the elongation $\rho(t) = s_0/s(t)$ is related to τ by

$$\tau \approx \frac{Dt}{s_0^2} \rho^2, \quad \text{for } \rho \gg 1 \quad 39.$$

and since $\theta \sim 1/\sqrt{\tau}$, the knowledge of the distribution of ρ in an ensemble of stretched lamella provides, via simple changes of variables, the distribution of τ (and therefore of t_s), and the distribution of θ (and therefore of c).

3.2.1. Sequential elongations: the lognormal paradigm. We consider protocols which consist in a sequential, uncorrelated (in intensity and direction) series of stretchings, applied either to a large collection of blobs, or to sub-parts of a stretching blob. This can be realized in several ways like in asymmetrical Baker transforms (Ott & Antonsen 1989) or other iterated maps (Meunier & Villiermaux 2010; Figueroa *et al.* 2014) and random processes (Kalda 2000), by the transport of the blob through successive pores in a porous medium (Le Borgne *et al.* 2015) or in a sheared suspension of beads (Souzy *et al.* 2017), for instance.

If the blob experiences N successive random stretchings ρ_i , its elongation is $\rho = \prod_i^N \rho_i$ and if the ρ_i 's are all independent, the probability $Q(\rho)$ that a point on the *initial* blob is stretched by a factor ρ is given by $Q(\rho) = \exp[-(\log \rho - N\mu)^2/2N\sigma^2]/\rho\sqrt{2\pi N\sigma^2}$, where μ and σ^2 are the mean and variance of $\log \rho$, defining a lognormal distribution. The probability $P(\rho)$ that a point on the *final* strip has experienced a stretching ρ is equal to $[s_0/\ell(t)]\rho Q(\rho)$ where $\ell(t)$ is the total length of the strip. With a number N of stretchings proportional to time t in a permanently stirred flow,

$$P(\rho) = \frac{s_0/\ell(t)}{\sqrt{4\pi\kappa t}} \exp \left[-\frac{(\log \rho - \gamma_p t)^2}{4\kappa t} \right] \quad 40.$$

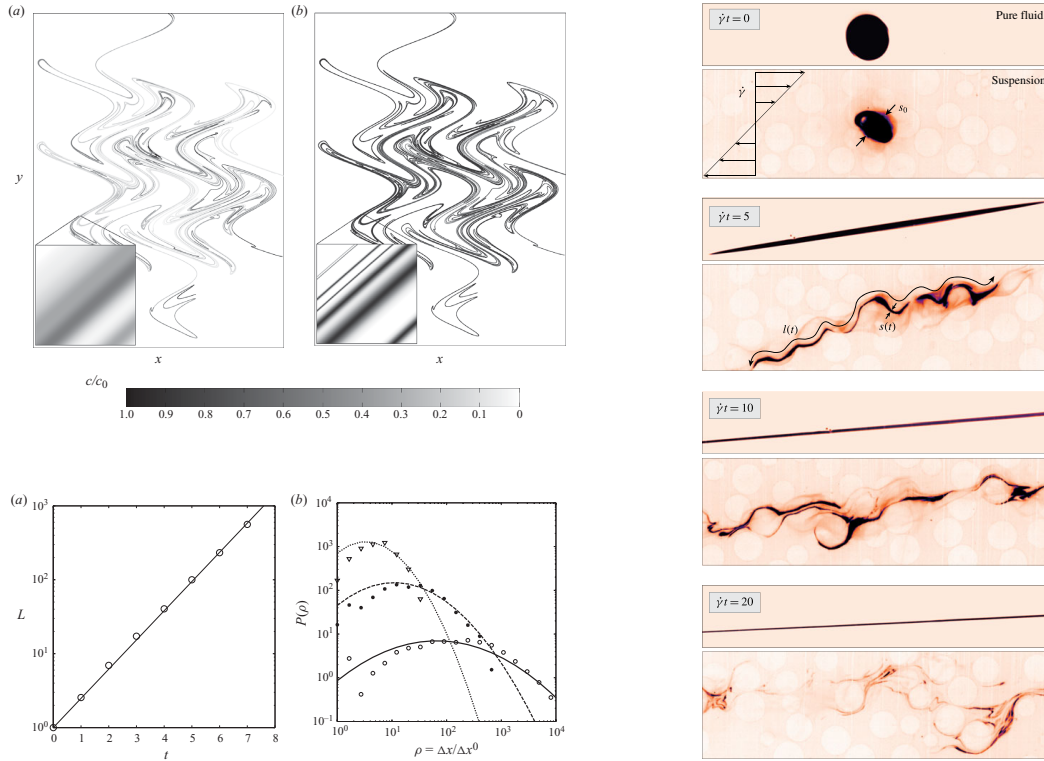


Figure 6

Sequential random stretchings and the lognormal paradigm: Mixing of a blob by a simulated Sine Flow (From Meunier & Villiermaux (2010)). A) The concentration field is reconstructed a-posteriori once the kinematics of the deformation and associated distribution of τ along the strip have been computed. The Péclet number $Pe = 10^7$ (left) and $Pe = 10^8$ (right) is then varied at will, show how lamella overlap earlier at lower Pe . B) Exponential growth of the lamella length $\ell(t)$ (a), and elongation distribution (b) represented by (40.) for three successive instants of time. C) Blob elongations in a sheared flow without (top), and in the presence of a suspension of beads (bottom) for successive instants of time. The heterogeneity of the local elongations of the strip is obvious (From Souzy *et al.* (2017)).

where $\gamma_p = N\mu/t$ is the most probable stretching rate, and $\kappa = N\sigma^2/(2t)$ stands for their dispersion, both depending of the type of unsteadiness in the flow (for instance Souzy *et al.* (2017) showed how γ_p and κ depend on the volume fraction of beads in a sheared suspension). The net length of the strip $\ell(t) = \int \rho Q(\rho) d\rho = s_0 e^{(\gamma_p + \kappa)t}$ increases exponentially fast, a common feature of random sequential processes (Cocke 1969; Hinch 1999; Duplat & Villiermaux 2000). **Figure 6** shows examples of this paradigm of sequential mixing, commonplace in real-world and numerical experiments. From $Q(\rho)$, apparent stretching rates $\gamma = (\ln \rho)/t$ (or finite-time Lyapunov exponents Bohr *et al.* (1998)) can be defined, whose distribution is

$$G(\gamma) = \sqrt{\frac{t}{4\pi\kappa}} e^{-\frac{(\gamma - \gamma_p)^2}{4\kappa} t} \quad 41.$$

showing how, as time proceeds, all elements of the strip experience progressively the same effective stretching given by the most probable stretching rate γ_p . Cumulated stretching histories are all alike as the mixture approaches uniformity. It was suggested by Kalda (see

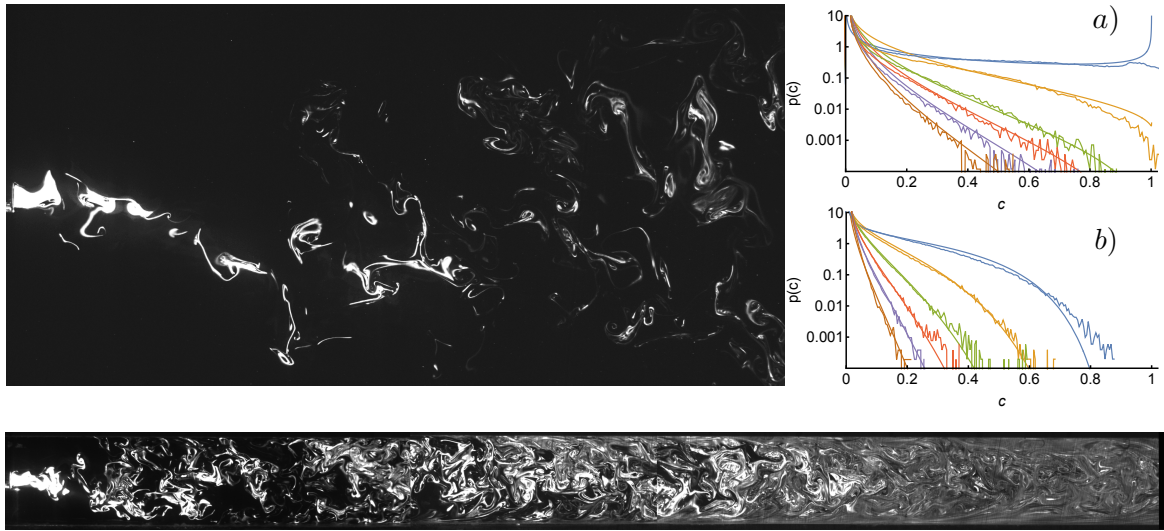


Figure 7

A) A planar cut through a dispersing plume made by the injection of a dye (Disodium Fluorescein in water) through a small tube $h = 4$ mm in diameter on the axis of a larger turbulent jet with $L = 8$ cm. Scalar sheets dilute by evolving on their own. B) Same as above, with the plume confined in a square duct of lateral width $L = 3$ cm. The mixture relaxes, through aggregation of sheets, towards a non-zero average concentration; $Re = 10^4$. a) Concentration distributions $p(c)$ measured at increasing distances from the source in A) as in Duplat *et al.* (2010a) and fits by (43.) for $Sc = 10^3$. b) Same as in a), with $Sc = 7$ (heat in water). From Le Borgne *et al.* (2017).

Meunier & Villermaux (2010)) that γ_p and κ should be related to each other by $\gamma_p = d\kappa$ in dimension d ; from the sole knowledge of the net growth rate of material surfaces, the entire distribution of elongations can be inferred.

The distributions $q(\theta)$ and $p(c)$ follow from $P(\rho)$ by quadratures (see 39.) and since θ , τ and ρ are power laws of each other in the long time limit, $p(c)$ is also lognormal (Le Borgne *et al.* 2015).

3.2.2. Solitary strips in turbulence. The multiplicative nature of the elongation process is such that strongly elongated portions of a blob are likely to be even more stretched in the next sequences. Since large elongation means short mixing time, the distribution of mixing times $T(t_s)$ obtained from (40.) with (21.) and (39.) is an essentially decaying function of t_s , well represented by (Shraiman & Siggia (1994), see also Duplat *et al.* (2010a))

$$T(t_s) = \frac{1}{\langle t_s \rangle} e^{-t_s / \langle t_s \rangle} \quad 42.$$

Crossover functions like $\theta \approx (1+t/t_s)^{-\beta}$ (Duplat *et al.* 2010a) or $\theta \approx 1 - e^{-(t_s/t)^\beta}$ (Le Borgne *et al.* 2017) are good fits for θ in (20.) leading, with (33.) to (Figure 7)

$$p(c) = \frac{\tilde{t}}{\beta} \frac{[-\log(1-c)]^{\frac{1}{\beta}-1}}{1-c} e^{-\tilde{t}[-\log(1-c)]^{\frac{1}{\beta}}}, \quad \text{with } \tilde{t} = \frac{t}{\langle t_s \rangle} \quad 43.$$

In the far field of a decaying turbulent jet with mean velocity u , the average mixing time of a solitary strip injected from a tube of diameter h smaller than the radius of the jet

is given by $\langle t_s \rangle \sim (h/u)Sc^{1/5}$, whose Schmidt number dependence was checked over three orders of magnitude. In a turbulent flow, the strip is chopped-off in lamella with thickness of the order of the Taylor scale $s_0 \sim \sqrt{\nu h/u}$, which are further stretched by the velocity gradient $\gamma \sim u/h$ at the scale of the injection tube according to $s(t) \sim s_0(\gamma t)^{-2}$, hence $Pe = \gamma s_0^2/D = Sc$ and $\beta = 5/2$ (Duplat *et al.* (2010a), see also Villermaux & Rehab (2000)). A similar construction involving exponentially distributed stretchings at each sequence of fixed duration $\langle t_s \rangle$ providing

$$p(c) = \frac{(-\ln c)^{\tilde{t}-1}}{\Gamma(\tilde{t})} \quad 44.$$

was proposed by Kalda & Morozenko (2008) in a slightly different context.

The exponentially decaying form of $p(c > \langle c \rangle) \sim e^{-\tilde{t}c}$ exemplifies rare events, getting rarer for increasing time, or distance to the source. These intermittent ‘still not yet mixed’ portions of the strip separated from each other by immense voids, carry a concentration way above the -otherwise close to zero- mean concentration $\langle c \rangle$ (Celani *et al.* 2014).

4. OVERLAPS

Solitary strips give a fair representation of the mixture composition as long as each of their sub-parts evolve on their own; but in most flows in practice, this lonely route has an end: a blob stretched exponentially in a bounded two-dimensional space occupies, after t_s , an area $s_0\sqrt{D/\gamma}e^{\gamma t}$ soon larger than the stirring domain. In turbulent flows, the strip gets corrugated, or ‘rough’ at all scales (i.e. fractal), with a fractal dimension depending both on scale (Catrakis & Dimotakis 1996), and time (Villermaux & Gagne 1994; Villermaux & Innocenti 1999; Nicolleau & Elmaihy 2004). The consequence of this inherent, or enforced confinement, is that a strip will unavoidably *overlap* with itself, and that the concentration levels along the strip is then no more that of an individual trajectory, but result from an interaction with neighboring portions of the strip (**Figure 8**).

4.1. Linearity of the Fourier equation: additions and convolutions

The Fourier equation (18.) is linear in c , and any concentration field $c(\xi, \tau)$ is the sum of Gaussian pulses with amplitude modulated by an appropriate initial condition $c(\xi, 0)$

$$c(\xi, \tau) = \int \frac{d\xi' c(\xi', 0)}{2\sqrt{\pi\tau}} e^{-\frac{(\xi-\xi')^2}{4\tau}} \quad 45.$$

Equivalently, a mixture is the sum of its sub-parts: the concentration profile of a set of two lamella 1 and 2 as those shown in **Figure 8**, each with a profile $c_1(\xi, \tau)$ and $c_2(\xi, \tau)$, is obtained by summation

$$c(\xi, \tau) = c_1(\xi, \tau) + c_2(\xi, \tau), \quad 46.$$

an elementary composition rule which is the building block of the evolution of complex mixtures. Indeed, if one divides a-priori a blob in two by tagging each sub-part with a different color and if $p_1(c_1)$ and $p_2(c_2)$ are the concentration distributions of each sub-fields, then the distribution $p(c)$ of the total concentration field $c = c_1 + c_2$ must be a combination of them.

For a broad variety of stirring protocols where the lamella are enforced to overlap, it has been found that additions in (46.) are made at random among the concentration levels

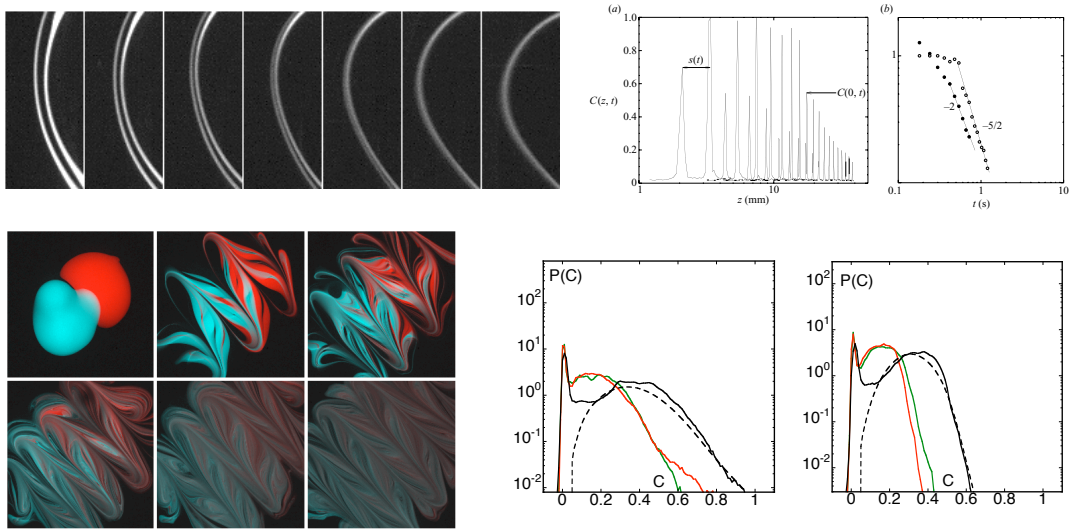


Figure 8

Lamella overlap and convolutions: A) Lamella aggregation and temporal evolutions of their separation distance $s(t) \sim t^{-2}$ and associated maximal concentration $\theta \sim t^{-5/2}$. B) Splitting of a blob in two different sub-parts (tagged in red and green), concentration distributions $p(c)$ of each distinct fields, of their convolution according to (47.) (dotted line), and of the sum of the two fields (black line), for two consecutive stirring cycles (From Duplat & Villermaux (2008); Duplat *et al.* (2010b)).

available in the current distributions. Random additions in concentration space translate into a convolution in probability space (Feller 1970)

$$p(c) = \int_{c=c_1+c_2} p_1(c_1)p_2(c_2)dc_2 = p_1 \otimes p_2 \quad 47.$$

and, when it actually succeeds in describing the mixture, gives a precise definition of what ‘random stirring protocol’ means. This is the case for interfering line sources (Warhaft 1984) and plumes (Duplat & Villermaux 2008) in turbulence, blobs stirred in viscous fluids (Duplat *et al.* 2010b), or porous media (Kree & Villermaux 2017). For these stirring protocols, all particles constitutive of the mixture have a chance to interact with all the others. This excludes flows with permanent segregated islands (Giona *et al.* 2004), or to some extent slow regions like near walls which prevent good blending (Gouillart *et al.* 2007).

4.2. Self convolution and Gamma distributions

Solitary strips evolve on their own in dispersing mixtures, but when confined, overlap according to (47.). The distribution $p(c, t + \delta t)$ is thus the result of a convolution with itself $p(c, t)$ an instant earlier necessary for the additions (46.) to complete. We confuse c and θ , and consider two limits, making use of the Laplace transform of $\tilde{p}(s, t) = \int_0^\infty p(c)e^{-sc}dc$.

- Either a fraction $r\delta t$ of the lamella, or sheets undergo a complete addition between t and $t + \delta t$, and in that case

$$\partial_t \tilde{p} = r(-\tilde{p} + \tilde{p}^2) \quad 48.$$

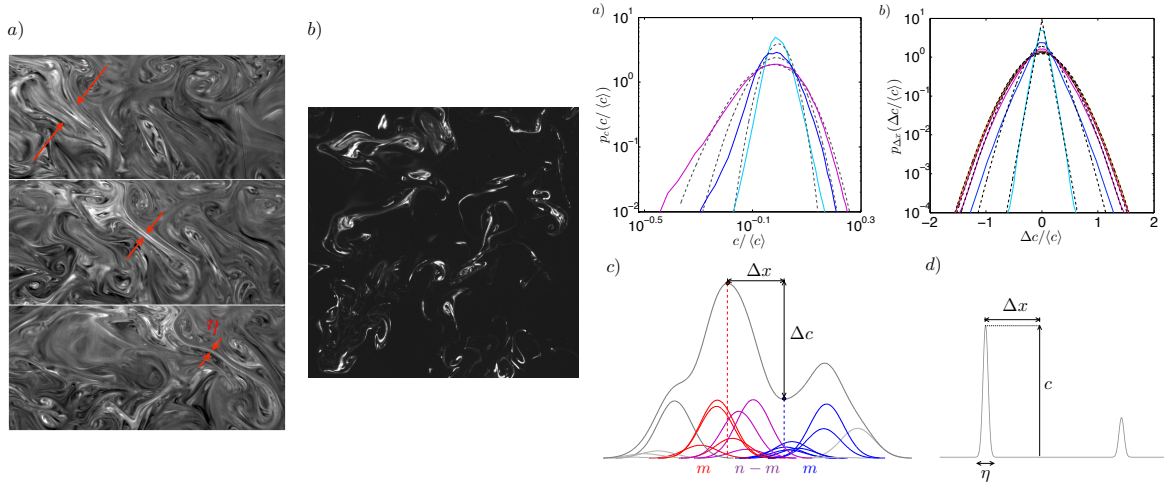


Figure 9

A) Three consecutive instants of times showing how a bundle of stretched sheets in a confined channel are brought together and merge, losing their individuality on a support of transverse thickness η , leading to Gamma distributions for $p(c)$ in (55.), and Bessel distributions for $p(\Delta c)$ in (62.). Close-up in the dispersing mixture showing how the scalar field resolves into a set of essentially non-interacting, disjoint sheets with distributed concentrations. c) Sketch of the elementary sheet overlapping mechanism constructing the concentration field: every concentration level c results from the merging of n sheets (the rapidly oscillating curves with concentrations θ) on a support of size η . Nearby concentration levels (separated by Δx) have in common the contribution of $n - m$ sheets. d) In dispersing mixtures where sheets are isolated and merging is anecdotal, concentration differences Δc are given by the concentration field c itself. From Le Borgne *et al.* (2017).

an equation familiar in the context of kinetic aggregation since von Smoluchowski (1917) (see also Curl (1963); Pope (1985); Pumir *et al.* (1991)), whose asymptotic solution is a decaying exponential irrespective of $p(c, 0)$, broadening in time

$$p(c, t) \sim \exp\left(-\frac{c}{e^{\int_0^t r dt'}}\right), \quad 49.$$

- Or, the convolution operation occurs on a *continuous* timescale everywhere in the flow, with sheets all merging with their neighbors in a continuous way, therefore altering the distribution $p(c, t)$ even on an infinitesimal timescale and in that case

$$\partial_t \tilde{p} = r \tilde{p} \ln \tilde{p} \quad 50.$$

whose solution is the self-convolution of the initial distribution $p(c, 0)$ as

$$p(c, t) = p(c, 0)^{\otimes \exp(\int_0^t r dt')}. \quad 51.$$

The two self-convolution routes above are distinct limits of the general evolution equation (Villermaux & Duplat 2003)

$$\partial_t \tilde{p} = rn \left(-\tilde{p} + \tilde{p}^{1+1/n} \right), \quad 52.$$

defining for $p(c, t)$ a *unique* family of distributions, with a single parameter n . The discrete time additions in (48.) corresponds to $n = 1$, and the uniform continuous time process in

(50.) is recovered when $n \rightarrow \infty$. To this crucial random addition step is superimposed the decay of c by stretching, resulting in a global shift $p(c + \delta c, t + \delta t) = p(c, t)$ with $\langle \delta c / \delta t \rangle = -\gamma(t)c$ so that the complete evolution of $p(c, t)$ is

$$\partial_t p = \gamma \partial_c (c p) + r n \left(-p + p^{\otimes 1+1/n} \right). \quad 53.$$

In mixtures with conserved average concentration, additions compensate for stretching, so that $r = \gamma$ and since the lamella aggregate *because* they are stretched, $\dot{n} = \gamma n$. The parameter n thus appears as a number of convolutions at time t and since the piling-up of the concentration levels by coalescence through (52.) contributes to a concentration increase $\exp\{\int dn/n\} = n$, the average concentration is conserved provided

$$n = \frac{1}{\theta(t)} \quad 54.$$

In that case, $p(c, t)$ is asymptotically given by

$$p(x = c/\langle c \rangle) = \frac{n^n}{\Gamma(n)} x^{n-1} e^{-nx}, \quad \text{with } \dot{n}/n = \gamma(t) \quad 55.$$

a Gamma distribution, with order $n(t)$ increasing in time, only function of the mixture rate of stretch while the shape of the distribution solely reflects the microscopic additions giving birth to it. Obviously, $p(x) \xrightarrow[n \rightarrow \infty]{} \delta(x - 1)$ when the mixture is completely *mixed* in the sense of (8.), and not simply blended as in (7.); it took the above developments to understand why and how.

This distribution represents well mixtures in turbulent channel flows (see Duplat & Villermaux (2008) and **Figure 9**), weakly heterogeneous porous media (Le Borgne *et al.* 2015), blobs in viscous fluids (Villermaux & Duplat 2003), along with their respective temporal dependence of n , specific to each stirring protocol. Numerical simulations have confirmed that the solitary strip concentration distribution has to be convoluted with itself $n(t)$ times to reconstruct the full overlapped mixture $p(c, t)$ (Meunier & Villermaux 2010; Le Borgne *et al.* 2015).

Additions in (46.) should be understood about the mean $\langle c \rangle$ and actually lead to the Gamma family (55.) when $\langle c \rangle \ll 1$. When $\langle c \rangle = 1/2$ for instance, the fluctuations of $c - \langle c \rangle$ are symmetrical about 0 and (53.) leads to Bessel functions (Villermaux *et al.* 2008).

4.3. Coarsening scale and Increments

The permanent lamella overlaps in a stirred mixture have a consequence not only on its concentration content, but also on its spatial structure.

4.3.1. Coarsening scale. To the balance between diffusion and stretching is associated a Batchelor scale (section 2.4); overlaps give rise to another, the coarsening scale (Villermaux & Duplat 2006). Sheets, lamella are typically dense in space in confined mixtures, and are locally parallel, aligned in the direction of stretching, forming bundles.

Consider for instance an initial scalar field $c(x, 0)$ consisting in a bundle of parallel lamella, each separated from their immediate neighbors by a distance s_0 , and piled-up over a distance of order L where the stretching applies: from $c(x, 0) = 1 + \cos(2\pi x/s_0)$ for $x \in \{-L/2, L/2\}$, we have $c(\xi, \tau) = 1 + \cos(\xi)e^{-\tau}$. The time needed to complete

the coalescence of the lamella in the bundle (**Figure 9**) is the time required to make the concentration modulations small compared to unity (i.e. $\tau > 1$). At that time t_s , the bundle where the merged lamella have percolated (Villermaux 2012a; Le Borgne *et al.* 2017) has shrunk down to the transverse size given by

$$\eta = LPe^{-\alpha/(2\alpha+1)} \quad 56.$$

for a power law stretching, or $\eta = LPe^{-1/2}$ for exponential stretching. Within η , which now scales as the stirring scale L and is thus much larger than the Batchelor scale, the concentration is close to uniform. This is the ‘scale of scrutiny’ imagined by Danckwerts (1953) to describe a mixture. This mechanism explains the ramp-cliff-plateau structures notorious in shear flows. There, L -wide regions of nearly uniform concentration are separated by steep cliffs absorbing a concentration difference of the order of the mean (Sreenivasan 1991; Pumir 1994; Warhaft 2000).

4.3.2. Increments: strips as quanta. In confined mixtures, diffusive overlap between elementary lamella occurs in bundles of transverse size η (section 4.3.1). There, the concentration $c(\mathbf{x})$ at a point \mathbf{x} results from the random superposition of concentration levels of n elementary strips, each of them with concentration θ_i as $c(\mathbf{x}) = \sum_{i=1}^n \theta_i(\mathbf{x}, t)$ leading for $p(c)$ to the self-convolution construction described in section 4.2. This aggregation mechanism has a consequence on the mixture spatial structure measured for instance by the distribution $p(\Delta c)$ of concentration increments $\Delta c(\Delta \mathbf{x}) = \langle c(\mathbf{x} + \Delta \mathbf{x}) - c(\mathbf{x}) \rangle$.

Consider two locations \mathbf{x} and $\mathbf{x} + \Delta \mathbf{x}$ separated by a distance $\Delta x < \eta$. There, the concentrations are $c(\mathbf{x}, t)$ and $c(\mathbf{x} + \Delta \mathbf{x}, t)$, respectively which both result from the addition of n independent levels θ_i so that

$$\Delta c(\Delta \mathbf{x}) = \sum_{i=1}^n \theta_i(\mathbf{x} + \Delta \mathbf{x}) - \sum_{i=1}^n \theta_i(\mathbf{x}), \quad 57.$$

the two sums being contributions from elementary lamella in a neighborhood of size η lying in \mathbf{x} , and $\mathbf{x} + \Delta \mathbf{x}$. Thus, when $\Delta x < \eta$, the two neighborhoods intersect, with $n - m$ lamella in the common overlapping region, and m independent lamella in the rest (**Figure 9**). Upon subtraction (57.), the levels from the $n - m$ lamella that contribute to both concentration levels cancel-out, and thus (Le Borgne *et al.* 2017)

$$\Delta c(\Delta x) = \sum_{i=1}^{n-m+m} \theta_i(\mathbf{x} + \Delta \mathbf{x}) - \sum_{i=1}^{n-m+m} \theta_i(\mathbf{x}) \quad 58.$$

$$= \sum_{i=1}^m \theta_i(\mathbf{x} + \Delta \mathbf{x}) - \sum_{i=1}^m \theta_i(\mathbf{x}) \quad 59.$$

$$= c'(\mathbf{x} + \Delta \mathbf{x}) - c'(\mathbf{x}), \quad 60.$$

where $c'(\mathbf{x} + \Delta \mathbf{x})$ and $c'(\mathbf{x})$ are now two independent concentrations obtained by random addition of $m \leq n$ independent lamella in the respective disjointed neighborhoods. Since the concentration levels $c'(\mathbf{x}, t)$ and $c'(\mathbf{x} + \Delta \mathbf{x}, t)$ are now statistically independent, the convolution rule (47.) applies, and

$$p(\Delta c) = \int dc' p(c'|m) p(|\Delta c| - c'|m). \quad 61.$$

where $p(c|m)$ is the concentration distribution of m aggregated independent lamella. Using (55.) with $\langle c \rangle = n\theta$, we have

$$p(\Delta c) = \frac{1}{\sqrt{\pi}\theta^{2m}\Gamma(m)} \left(\frac{|\Delta c|\theta}{2}\right)^{m-1/2} K_{m-1/2}\left(\frac{|\Delta c|}{\theta}\right), \quad 62.$$

The number m obviously increases with the separation distance Δx , and $m \rightarrow n$ as $\Delta x \rightarrow \eta$. Equation (61.) illustrates how computing increments of concentration in a field made of elementary aggregations *deconstructs* the direct aggregation process. One probes all the more deep, or early, in the process that small scale increments are considered since the number of independent lamella vanishes as $\Delta x \rightarrow 0$. When $m \rightarrow 1$, $p(c|1)$ is a measure of the ‘quantum’ (Villermaux 2012a), or elementary brick constructing the concentration field $p(c)$, that is the solitary strip. The spatial correlation of the concentration field in a confined mixture thus results from an uncorrelated, random superposition of quanta, or strips. Their possible entanglement (Duplat *et al.* 2010b) singles-out long lasting temporal correlations from the mixture initial condition and/or stirring protocol.

SUMMARY POINTS

1. Mixing is not blending, nor stirring. Mixing is stretching enhanced diffusion and results from a subtle interplay between substrate deformation, and diffusion broadening when the Péclet number is large.
2. In most instances involving mixing, the interest is in controlling the probability of occurrence of large, or low concentrations: There is therefore a need to understand the concentration distributions $p(c)$, and especially their large excursion tails.
3. Concentrations c are described by a pure diffusion equation $\partial_\tau c = \partial_\xi^2 c$ in suitably chosen variables $\{\xi, \tau\}$ function of the nature of the local stretch history, and of molecular diffusion. They decay after a *mixing time* $t_s \sim \gamma^{-1}\mathcal{F}(Pe)$, essentially fixed by the deformation rate of the substrate γ , corrected by a (usually weak, but singular) function of the Péclet number Pe depending on the stirring protocol.
4. Stirring motions form lamella typically unevenly stretched. When these solitary strips evolve on their own, they carry a distribution of concentration $p(c)$ reflecting the elongation histories along the strip only.
5. In confined mixtures where the strips are enforced to overlap, concentration levels add at random and $p(c)$ is now constructed by a self-convolution rule setting its shape (a Gamma distribution), and directing its evolution towards uniformity.
6. Bundles of lamella aggregate at the coarsening lengthscale η , larger than the Batchelor scale, proportional to the stirring scale, and to a decaying function of Pe . Within a range of scales $\Delta x \leq \eta$, the distribution of concentrations differences $p(\Delta c)$ is a deconstruction the direct aggregation process giving birth to $p(c)$.

FUTURE ISSUES

1. Heuristics: The present ideas and methods are not limited to passive scalars. They were successfully applied to evaporating dense sprays (Villermaux *et al.* 2017), Marangoni flows (Geri *et al.* 2017; Néel & Villermaux 2018), and should contribute

to reexamine old problems like mixing in stratified flows (Osborn 1980) or in chemically reactive mixtures (Gibson & Libby 1972; Tél *et al.* 2005), to the “demixing” of colloids by diffusiophoresis (Prieve *et al.* 1984; Deseigne *et al.* 2014; Shin *et al.* 2017; Raynal *et al.* 2018), and could be applied to mixing by living animals (biomixing, see e.g. Kurtuldu *et al.* (2011)), or by optimized stirring protocols (Thiffeault 2012; Weij & Bartolo 2017), among other fascinating topics.

2. Fundamentals: In confined mixtures, the self-convolution route towards uniformity is an empirical fact. However, the status of this ubiquitous ‘maximal randomness’ property of random flows is unclear. A simple case using maps could be worked-out to understand the decay of correlation of τ in space (hence ensuring the independence of concentrations at merging); that might not be a simple exercise (Gilbert 2006), although certainly a useful one.
3. Numerics: A solitary strip carries concentrations reflecting its elongation history (i.e. τ) only. This fact has prompted the Diffusive Strip Method (DSM) a simulation method to compute a-posteriori a mixture from the kinematics of the flow, for any $Pe > 1$. Working both in 2-d (Meunier & Villermaux 2010) and 3-d (Martinez-Ruiz *et al.* 2018), it could have a broad range of applications.

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