Nonuniform mixing

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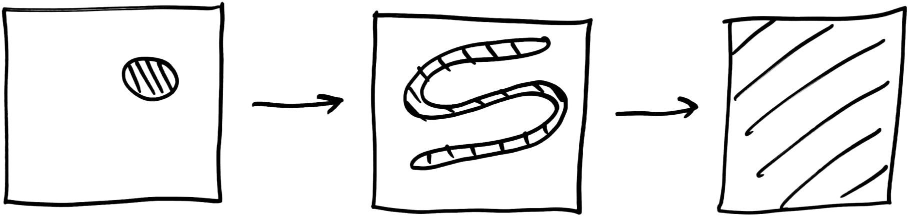
Uniform mixing

The usual scenario in mixing is that we want to homogenize some initial distribution of particles or dye.



This will happen naturally via molecular diffusion, but is greatly accelerated by stirring.

See for instance Welander (1955); Young (1999); Doering & Nobili (2020).

Mathematical description

The advection-diffusion equation governs the evolution of a passive scalar concentration $\theta(\boldsymbol{x}, t)$:

$$\partial_t \theta + \boldsymbol{u} \cdot \nabla\theta = D\,\nabla^2\theta, \quad \nabla \cdot \boldsymbol{u} = 0 \quad \text{in } \Omega$$

where $\boldsymbol{u}(\boldsymbol{x}, t)$ is a divergence-free velocity field, and D is the diffusivity.

We typically use no-flux boundary conditions

$$\boldsymbol{F}[\theta] \cdot \hat{\boldsymbol{n}} := (\boldsymbol{u}\,\theta - D\nabla\theta) \cdot \hat{\boldsymbol{n}} = 0 \quad \text{on } \partial\Omega \text{ (boundary).}$$

The integral $\int_\Omega \theta\,\mathrm{d}V$ is conserved:

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_\Omega \theta\,\mathrm{d}V = -\int_\Omega \nabla \cdot \boldsymbol{F}[\theta]\,\mathrm{d}V = -\int_{\partial\Omega} \boldsymbol{F}[\theta] \cdot \hat{\boldsymbol{n}}\,\mathrm{d}S = 0.$$

Eventually everything is mixed

How do we know that the concentration will eventually mix? A few integration by parts and use of boundary conditions give

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_\Omega \theta^2 \,\mathrm{d}V = -2D\int_\Omega |\nabla\theta|^2 \,\mathrm{d}V \le 0.$$

The decay of variance (L^2 norm) is monotonic: it can never increase. It can only stop decreasing if θ is uniform in space ($\nabla\theta \equiv 0$).

This bound underpins the usefulness of variance as a measure of mixing.

It is also useful in rigorous math as an *a priori* estimate that must be satisfied if strong solutions exist.

Compressible mixing

What if the velocity field is compressible? Then the fluid density $\rho(\boldsymbol{x},t) > 0$ is solved for along with the concentration:

$$\partial_t \rho + \nabla \cdot (\boldsymbol{u}\rho) = 0, \qquad \partial_t(\rho\,\theta) + \nabla \cdot (\boldsymbol{u}\rho\,\theta) = D\,\nabla^2\theta.$$

Notice that $\theta =$ const. is still a solution, so the ultimate steady state remains uniform.

The concentration variance equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_\Omega \rho\,\theta^2\,\mathrm{d}V = -2D\int_\Omega |\nabla\theta|^2\,\mathrm{d}V \leq 0,$$

again assuming no-flux boundary conditions on θ.

The variance will decay to zero over time, implying that $\theta(\boldsymbol{x},t)$ reaches the uniform mixed state.

In that sense compressible mixing is also an instance of a uniform mixing scenario.

Nonuniform mixing

The relaxation to a uniform state requires this uniform state to be a steady solution of the advection–diffusion equation.

This is not always the case!

Consider again:

$$\partial_t \theta + \nabla \cdot \boldsymbol{F}[\theta] = 0, \quad \boldsymbol{F}[\theta] = \boldsymbol{u}\,\theta - D\nabla\theta, \quad \text{in } \Omega$$

with

$$\boldsymbol{F}[\theta] \cdot \hat{\boldsymbol{n}} = 0 \quad \text{on } \partial\Omega.$$

But now we assume neither $\nabla \cdot \boldsymbol{u} = 0$ nor $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} = 0$ on $\partial\Omega$, the latter of which was a hidden assumption so far.

Uniform steady state

If we insert $\theta(\boldsymbol{x}, t) = \theta_0 =$ const. into our equation, we get

$$\theta_0 \nabla \cdot \boldsymbol{u} = 0 \quad \text{in } \Omega$$

and for the boundary conditions

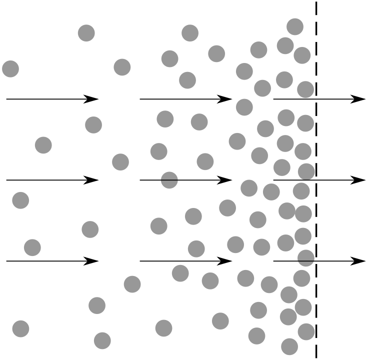
$$\boldsymbol{F}[\theta_0] \cdot \hat{\boldsymbol{n}} = \theta_0 \, \boldsymbol{u} \cdot \hat{\boldsymbol{n}} = 0 \quad \text{on } \partial\Omega.$$

We thus see the necessity of both $\nabla \cdot \boldsymbol{u} = 0$ and $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} = 0$ for the existence of a uniform steady state.

If either condition is violated, then the steady state is nonuniform. In fact it may not be steady at all.

Example: Particle filter

The simplest example of a nonuniform steady state is a filter: $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} \neq 0$ at the boundary, since fluid can cross the filter, but the particles cannot.



The equilibrium state is then nonuniform: particles tend to accumulate at suction regions on the boundary.

Example: Particle filter (cont'd)

A simple one-dimensional model for a filter has domain $\Omega = [0, L]$ and velocity $\boldsymbol{u} = U\,\hat{\boldsymbol{x}}$:

$$\partial_t\theta + U\partial_x\theta - D\,\partial_x^2\theta = 0, \qquad 0 < x < L$$

with no-flux boundary conditions at 0 and L:

$$U\theta - D\,\partial_x\theta = 0, \qquad x = 0, L.$$

Here $\nabla\cdot\boldsymbol{u} = 0$ but $\boldsymbol{u}\cdot\hat{\boldsymbol{n}}|_{\partial\Omega} = \pm U \neq 0$. The steady state is

$$\varphi(x) = \frac{U}{D}\,\frac{\mathrm{e}^{Ux/D}}{\mathrm{e}^{UL/D} - 1}\,.$$

The flow pushes particle against the boundary at $x = L$ (for $U > 0$), creating a boundary layer of thickness D/U.

Example: Surfactants

The equation for a surfactant concentration $\theta(\boldsymbol{x}, t)$ evaluated at a free surface is (Aris, 1989; Stone, 1990)

$$\partial_t \theta + \nabla_{\rm s} \cdot (\boldsymbol{u}_{\rm s}\, \theta) = D\, \nabla_{\rm s}^2 \theta - \theta\, (\nabla_{\rm s} \cdot \hat{\boldsymbol{n}}_{\rm s})\, \boldsymbol{u} \cdot \hat{\boldsymbol{n}}_{\rm s}$$

The surface velocity $\boldsymbol{u}_{\rm s}$ is not generally divergence-free. Surfactants can collect at 'downwelling' regions, in a similar manner to drifters in the ocean.

Eexample: Microswimmers

In a popular model for 2D swimmers, the probability density $p(\boldsymbol{x}, \phi, t)$ obeys a Fokker–Planck (or Smoluchowski) equation

$$\partial_t p + (\boldsymbol{u} + U\hat{\boldsymbol{q}}) \cdot \nabla p = D\,\nabla^2 p + D_{\text{rot}}\,\partial_\phi^2 p$$

with direction of swimming $\hat{\boldsymbol{q}} = (\cos\phi, \sin\phi)$ and rotational diffusion D_{rot}.

The fluid velocity $\boldsymbol{u}(\boldsymbol{x}, t)$ obeys $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} = 0$ at boundaries, but the swimming velocity $U\hat{\boldsymbol{q}}$ does not: a particle may keep pushing against a boundary even after it makes contact. (It is prevented from entering the wall by the no-flux boundary condition on p.)

Hence, the steady solution is not uniform: swimmers tend to accumulate near boundaries, in a manner similar to the filter example [Lee (2013); Ezhilan & Saintillan (2015); Chen & Thiffeault (2021)].

Example: Evaporation and precipitation

Concentration $\theta(t,z)$ of a passive scalar (e.g., salt) confined to time-varying domain $\Omega(t) = [0, h(t)]$:

$$\begin{aligned} \partial_t \theta - D_0\, \partial_z^2 \theta &= 0, & z &\in (0, h(t)); \\ \partial_z \theta &= 0, & z &= 0, \\ \dot{h}\,\theta + D_0\, \partial_z \theta &= 0, & z &= h(t). \end{aligned}$$

No scalar flux at either the top or bottom.

Physically, this models rain and evaporation on the surface of a body of water, with $\dot{h} < 0$ corresponding to evaporation, and $\dot{h} > 0$ corresponding to rain.

[Part of ongoing work with Albion Lawrence and Raf Ferrari.]

Example: Evaporation and precipitation (cont'd)

Since time-dependent boundaries are tricky to deal with, we rescale the variables as

$$Z = z/h(t), \qquad \psi(t,Z) = h\,\theta(t,hZ)$$

and obtain the advection-diffusion equation

$$\begin{aligned} \partial_t\psi + \partial_Z(W\psi) - D\,\partial_Z^2\psi &= 0, \qquad & Z &\in (0,1);\\ \partial_Z\psi &= 0, & Z &= 0,\\ W\psi - D\,\partial_Z\psi &= 0, & Z &= 1, \end{aligned}$$

with time-dependent velocity and diffusion:

$$W(t,Z) = -Z\dot{h}(t)/h(t), \qquad D(t) = D_0/h^2(t).$$

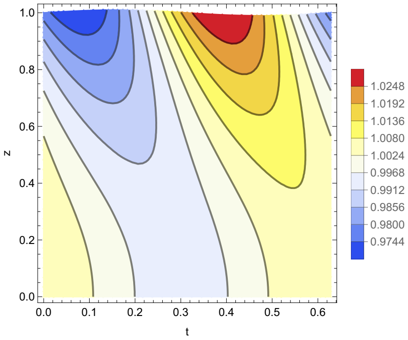
There is an 'apparent velocity' due to the moving surface. The velocity does not vanish at the top surface, so $\boldsymbol{u}\cdot\hat{\boldsymbol{n}} \neq 0$.

Example: Evaporation and precipitation (cont'd)

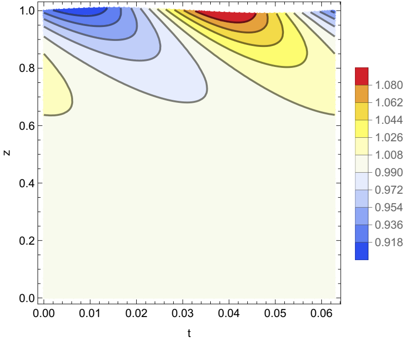
There is no steady solution to this problem. Rather, for periodic $h(t) = h_0 + H\sin(\omega t)$ all solutions converges to a periodic limit $\varphi(z,t)$.

$$\varepsilon = H/h_0 = 0.01$$

$$\omega h_0^2/D = 10$$



$$\omega h_0^2/D = 100$$



Relaxation to equilibrium

So what's the big deal? It still makes sense to define concentration variance as

$$\int_\Omega |\theta - \varphi|^2 \, \mathrm{d}V$$

where φ is the ultimate state (not necessarily uniform or steady).

With $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} \neq 0$ on the boundary, the evolution of variance is now given by

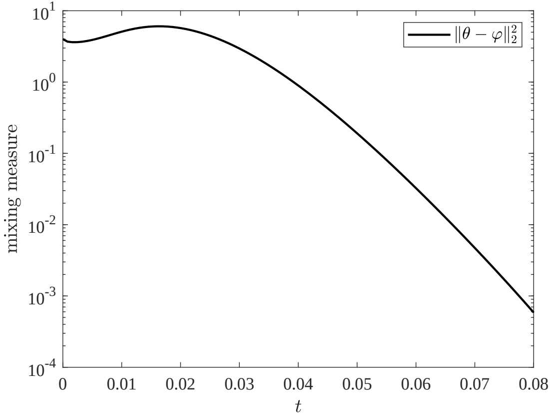
$$\frac{\mathrm{d}}{\mathrm{d}t} \int_\Omega |\theta - \varphi|^2 \, \mathrm{d}V = \boxed{\int_{\partial\Omega} |\theta - \varphi|^2 \, \boldsymbol{u} \cdot \hat{\boldsymbol{n}} \, \mathrm{d}S} - 2D \int_\Omega |\nabla(\theta - \varphi)|^2 \, \mathrm{d}V.$$

Note the boundary term on the right is not sign-definite. Hence variance no longer has to decrease monotonically. It can exhibit transient growth.

Of course variance must ultimately decay, which we know from other considerations. But the above equation does not show that, and suggests that variance can be poorly-behaved if used as a measure of mixing.

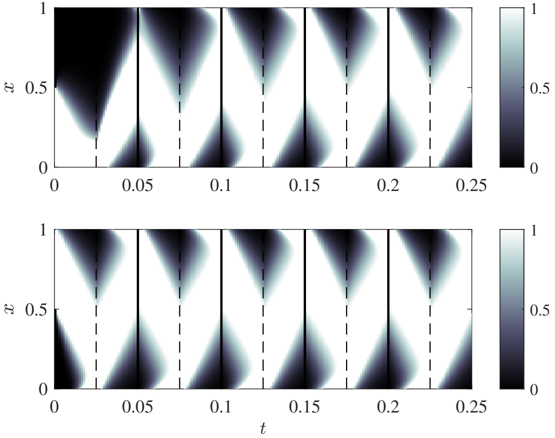
Relaxation to equilibrium: Example

In practice the variance can transiently increase, for instance in our uniform flow example:



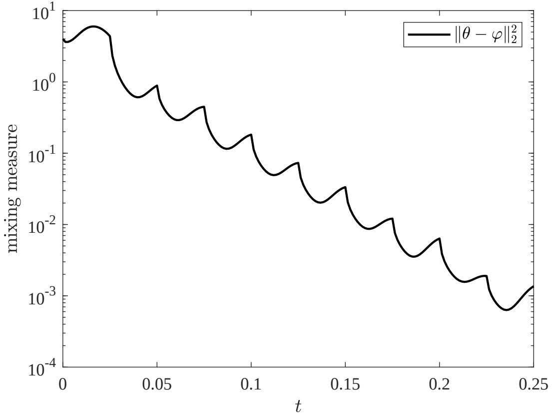
Relaxation to periodic solution: Example

We periodically reverse the direction of the flow ('breathing').



Two different initial conditions converge to the same periodic solution $\varphi(\boldsymbol{x}, t)$.

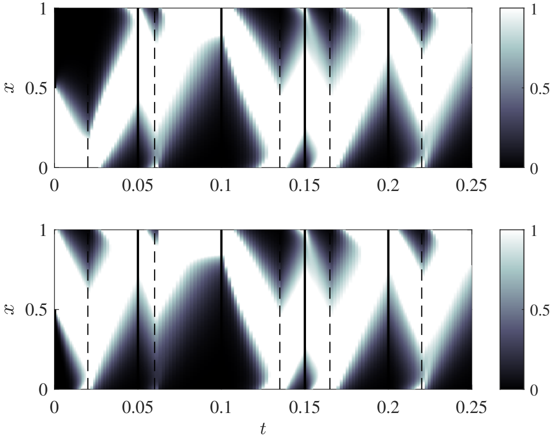
Relaxation to periodic solution: Convergence



Notice that variance (solid line) shows significant oscillations. Fitting a decay rate to this, or trying to optimize, is more challenging than it needs to be.

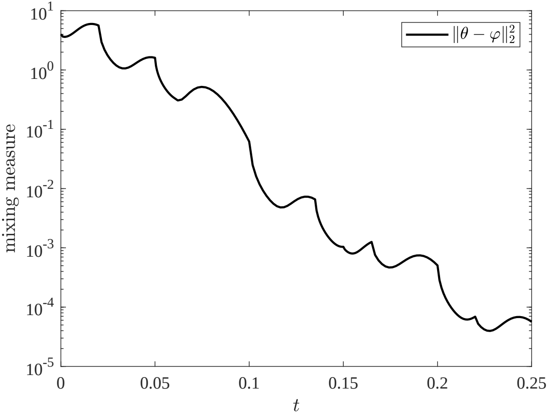
Relaxation to aperiodic solution: Example

We reverse the direction of the flow at random times.



Two different initial conditions converge to the same aperiodic solution $\varphi(\boldsymbol{x}, t)$.

Relaxation to aperiodic solution: Convergence



Again nonmonotonic.

How can we improve this?

To summarize so far:

- When either $\nabla \cdot \boldsymbol{u} \neq 0$ or $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} \neq 0$, the ultimate state φ is not uniform.
- The ultimate state can be steady, periodic, or aperiodic, depending on the time dependence of $\boldsymbol{u}(\boldsymbol{x}, t)$ and $D(\boldsymbol{x}, t)$.
- The mixing rate should be defined as the rate of convergence of any initial condition to $\varphi(\boldsymbol{x}, t)$.
- Alternatively, we can define the mixing rate as the rate of convergence of any two initial conditions to each other.
- However, the L^2 norm (variance) is not monotonically decreasing, which is undesirable.
- Can we improve this by using a different mixing measure?

Entropy and f-divergence

An alternative measure of mixing in the nonuniform case is the f-divergence (Österreicher & Vajda, 2003; Liese & Vajda, 2006):

$$H_f[p_1, p_2] := \int_\Omega p_2 \, f(p_1/p_2) \, \mathrm{d}V.$$

Here p_1, $p_2 \geq 0$ are two normalized probability densities, and f is a convex function with $f(1) = 0$, $f'' > 0$.

For example we can choose

$$f(u) = u \log u$$

which gives the Kullback–Leibler divergence or relative entropy.
[Note: opposite sign of physics convention.]

H_f measures the 'distance' (divergence) between p_1 and p_2. Not in general symmetric, so not a metric!

Time-evolution of f-divergence

Take p_i to satisfy the advection-diffusion problem:

$$\partial_t p_i = -\nabla \cdot \boldsymbol{F}[p_i], \qquad \boldsymbol{F}[p_i] = \boldsymbol{u}\, p_i - \mathbb{D} \cdot \nabla p_i$$

with $\boldsymbol{F}[p_i] \cdot \hat{\boldsymbol{n}} = 0$ on $\partial\Omega$.

$$\begin{aligned}
\dot{H}_f[p_1, p_2] &= \frac{\mathrm{d}}{\mathrm{d}t} \int_\Omega p_2\, f(p_1/p_2)\, \mathrm{d}V \\
&= \int_\Omega \big(\partial_t p_2\, f(p_1/p_2) + p_2\, f'(p_1/p_2)\big(\partial_t p_1/p_2 - p_1\, \partial_t p_2/p_2^2\big)\big)\, \mathrm{d}V \\
&= -\int_\Omega \Big(\nabla \cdot \boldsymbol{F}[p_2]\, f(p_1/p_2) \\
&\qquad + f'(p_1/p_2)(\nabla \cdot \boldsymbol{F}[p_1] - (p_1/p_2)\nabla \cdot \boldsymbol{F}[p_2])\Big)\, \mathrm{d}V.
\end{aligned}$$

We integrate by parts, and two terms containing $f'(p_1/p_2)\, \boldsymbol{F}[p_2] \cdot \nabla(p_1/p_2)$ cancel.

Time-evolution of f-divergence (cont'd)

We are left with

$$\dot{H}_f[p_1, p_2] = \mathsf{BT}[p_1, p_2] + \int_\Omega p_2^{-1} f''(p_1/p_2) \nabla(p_1/p_2) \cdot (p_2 \boldsymbol{F}[p_1] - p_1 \boldsymbol{F}[p_2]) \, \mathrm{d}V$$

The boundary terms vanish when $\boldsymbol{F}[p_i] \cdot \hat{\boldsymbol{n}} = 0$ on $\partial\Omega$.

Crucially:

$$\begin{aligned} p_2 \boldsymbol{F}[p_1] - p_1 \boldsymbol{F}[p_2] &= p_2(\boldsymbol{u}\, p_1 - \mathbb{D} \cdot \nabla p_1) - p_1(\boldsymbol{u}\, p_2 - \mathbb{D} \cdot \nabla p_2) \\ &= -p_2 \mathbb{D} \cdot \nabla p_1 + p_1 \mathbb{D} \cdot \nabla p_2 \\ &= -p_2^2 \, \mathbb{D} \cdot \nabla (p_1/p_2). \end{aligned}$$

Relaxation of f-divergence

We finally obtain the time-evolution

$$\boxed{\frac{\mathrm{d}}{\mathrm{d}t} H_f[p_1, p_2] = -\int_\Omega p_2\, f''(p_1/p_2)\, \nabla(p_1/p_2) \cdot \mathbb{D} \cdot \nabla(p_1/p_2)\, \mathrm{d}V \leq 0}$$

for general no-flux boundary conditions, that is, even if $\boldsymbol{u} \cdot \hat{\boldsymbol{n}} \neq 0$. The relaxation of f-divergence is thus always monotonic.

This is essentially an H-theorem from statistical physics. The novelty here is that in those applications the boundary conditions are often not at the forefront, since quantities such as momentum vanish at infinity. In the fluid-dynamical context it is precisely the no-flux boundary conditions that give this monotonic evolution of H_f.

Choice of $f(u)$

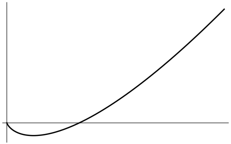
The convex function f is so far arbitrary:

$$H_f[p_1, p_2] = \int_\Omega p_2\, f(p_1/p_2)\,\mathrm{d}V.$$

There are many good choices, with different trade-offs.

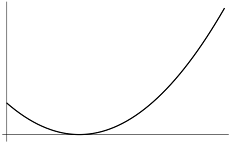
Relative entropy: $f(u) = u \log u$:

$$H_{\mathrm{KL}}[p_1, p_2] := \int_\Omega p_1\, \log(p_1/p_2)\,\mathrm{d}V.$$



Chi-square divergence: $f(u) = (1-u)^2$:

$$H_\chi[p_1, p_2] := \int_\Omega \frac{(p_1 - p_2)^2}{p_2}\,\mathrm{d}V.$$

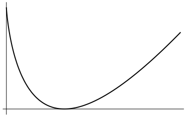


Almost like concentration variance, but not quite!

Choice of $f(u)$ (cont'd)

Jensen–Shannon divergence:

$$f(u) = \tfrac{1}{2} u \log u - \tfrac{1}{2}(1+u)\log[\tfrac{1}{2}(1+u)]$$

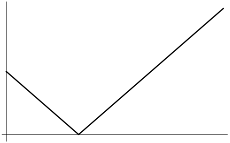


$$\begin{aligned} H_{\mathrm{JS}}(p_1, p_2) &= \tfrac{1}{2}\{H_{\mathrm{KL}}(p_1, p_{12}) + H_{\mathrm{KL}}(p_2, p_{12})\} \\ &= \tfrac{1}{2}\int_\Omega \{p_1 \log(p_1/p_{12}) + p_2 \log(p_2/p_{12})\}\,\mathrm{d}V \end{aligned}$$

where $p_{12} := \tfrac{1}{2}(p_1 + p_2)$. $\sqrt{H_{\mathrm{JS}}}$ is a metric, and it is bounded:

$$H_{\mathrm{JS}}(p_1, p_2) \le \tfrac{1}{2}(\log 2)\,\|p_1 - p_2\|_1 \le \log 2.$$

Total variation distance: $f(u) = \tfrac{1}{2}|1-u|$:



$$H_{\mathrm{TV}}[p_1, p_2] = \tfrac{1}{2}\int_\Omega |p_1 - p_2|\,\mathrm{d}V \le 1.$$

This is just the L^1 norm!

L^1 norm

The L^1 norm ($f(u) = |1-u|$)

$$\int_\Omega |p_1 - p_2| \,\mathrm{d}V = \int_\Omega |\theta| \,\mathrm{d}V, \qquad \theta := p_1 - p_2$$

is an f-divergence, unlike L^2.

However, the time-evolution from earlier requires f' and f'', which are not defined at 0.

We can derive a formula customized for L^1:

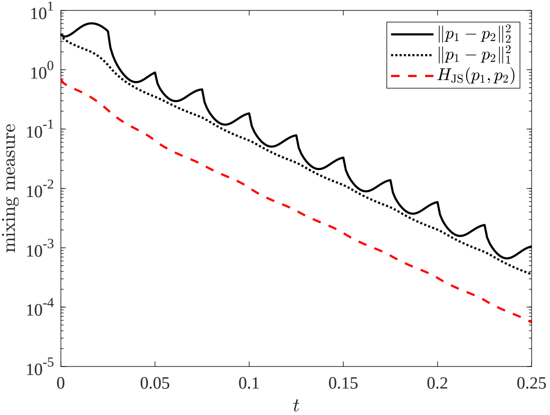
$$\boxed{\frac{\mathrm{d}}{\mathrm{d}t}\int_\Omega |\theta| \,\mathrm{d}V = -2\int_{\{\theta=0\}} \nabla\theta \cdot \mathbb{D} \cdot \nabla\theta \,\frac{\mathrm{d}S}{|\nabla\theta|} \le 0}$$

where the integral on the right is taken over the zero level set of $\theta(\cdot, t)$.

This suggests that L^1 is a more reliable measure of mixing than variance for nonuniform mixing.

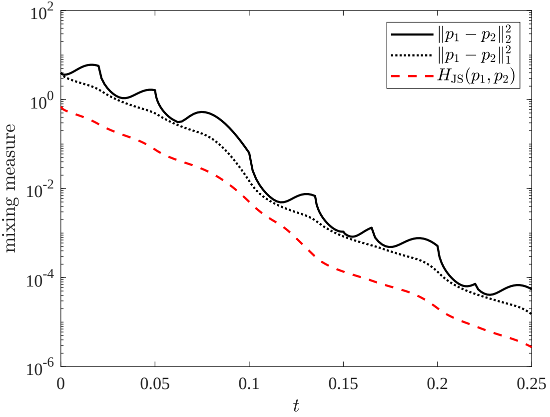
Relaxation of f-divergence: Periodic example

Return to the earlier periodic flow example: the dashed red line is the f-divergence H_{JS}. Notice how nice and monotonic it is compared to variance (solid).



Relaxation of f-divergence: Aperiodic example

Also true in the aperiodic case:



Generalization to number density

For the f-divergence, we require the passive scalar p to be a normalized probability distribution.

We can generalize the formalism to a number density $n(\boldsymbol{x}, t)$:

$$\partial_t n + \nabla \cdot \boldsymbol{F}(n) = Q(\boldsymbol{x}, t; n), \qquad \boldsymbol{x} \in \Omega.$$

The number of particles

$$N(t) = \int_\Omega n(\boldsymbol{x}, t)\, \mathrm{d}V$$

can change in time:

$$\dot{N} = \int_\Omega Q\, \mathrm{d}V.$$

Sources and sinks

We can't take the source-sink Q to be general, since we need to maintain $n(\boldsymbol{x}, t) \geq 0$. A common, sensible form is

$$Q(\boldsymbol{x}, t; n) = S(\boldsymbol{x}, t) - K(\boldsymbol{x}, t)\, n, \qquad S \geq 0,\ K \geq 0.$$

With this choice of source-sink, we make the obvious generalization

$$H_f[n_1, n_2] = \int_\Omega n_2\, f(n_1/n_2)\, \mathrm{d}V$$

which is no longer strictly-speaking an f-divergence. We make the further trivial restriction that $f \geq 0$.

Time evolution

We find after a similar process as earlier:

$$\dot{H}_f[n_1, n_2] = -\int_\Omega n_2\, f''(n_1/n_2)\,\nabla(n_1/n_2)\cdot\mathbb{D}\cdot\nabla(n_1/n_2)\,\mathrm{d}V \\ -\int_\Omega (Kn_2 f(n_1/n_2) + S\,g_f(n_1/n_2))\,\mathrm{d}V \leq 0 \quad (*)$$

where

$$g_f(u) := (u-1)f'(u) - f(u) \geq 0, \qquad g_f(1) = 0. \qquad (**)$$

The inequality in (*) follows from the positivity of n_i, the strict convexity of f ($f'' > 0$), the positive-definiteness of $\mathbb{D}$, the nonnegativity of f, K, S, and the inequality in (**).

The right-hand side of (*) vanishes if and only if $n_1 = n_2$.

Conclusions

- Mixing is usually regarded as the relaxation to a uniform state.
- The concentration variance (L^2 norm) is often taken as a convenient measure, since it relaxes monotonically to a uniform state.
- However, in some cases the ultimate state is not uniform.
- For example: suction boundary conditions, or divergent flows.
- In those nonuniform cases variance is less reliable, since it can exhibit oscillations: it is not constrained to decay monotonically.
- Better measures of mixing in the nonuniform case are the entropy-like quantities called f-divergences, including the L^1 norm.
- Since we usually don't know the ultimate state $\varphi(\boldsymbol{x}, t)$, it is preferable to look for convergence of two different initial conditions [see atmospheric tracers in Haynes & Shuckburgh (2000)].
- See Thiffeault, J.-L. (2021). *Physical Review Fluids,* **6** (9), 090501.

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