# The Development of Chaotic Advection

Hassan Aref
*Department of Theoretical and Applied Mechanics, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA*

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The concept was developed some twenty years ago as an outgrowth of work on advection by interacting point vortices. The term 'chaotic advection' was first introduced in the title of an abstract for the 35$^{th}$ annual meeting of the APS Division of Fluid Dynamics (DFD) in 1982. The main reference, a *Journal of Fluid Mechanics* paper published in 1984, may be the true 'birthdate' of the term. Earlier work from the 1960s by Arnol'd and Hénon on advection by steady 3D flows already contained closely related ideas and results but was not widely appreciated. The present paper, based on the 2000 Otto Laporte Memorial Lecture delivered at the 53$^{rd}$ APS/DFD annual meeting, traces these and other precursors and the development of chaotic advection over the past two decades. Some exciting recent developments, such as application to fluid mixing in MEMS, and to materials processing, and the introduction of topological methods of analysis, are highlighted. On balance, chaotic advection is now established as a subtopic of fluid mechanics with wide ramifications and continued promise for theory, experiment and applications.

## 1. INTRODUCTORY REMARKS

First, I should like to thank the Committee for awarding me the Otto Laporte Award for the year 2000. Next, I should like to thank those who nominated me for this award. Finally, I want to thank my collaborators and students over the years who made the nomination possible and viable. Thanks to you all. I greatly appreciate being selected.

The concluding sentence of the nomination reads: "...and most notably for the development of the concept of chaotic advection." So, although this development took place some two decades ago now, I thought the most appropriate topic for my lecture would, indeed, be the development of chaotic advection.

I also thought it appropriate to try to connect to Otto Laporte in my lecture. Otto Laporte was a member of the small group of brilliant, young theoretical physicists who received their training during the mid-1920's under Arnold Sommerfeld in Münich. Born in Mainz, Germany, on July 23, 1902, Laporte began doing experiments with optics and electromagnetism already in grade school. His first university courses were taken in Frankfurt where Max Born was among his professors. Born enthusiastically recommended him to Sommerfeld. In the group around Sommerfeld Laporte met and worked with Werner Heisenberg, Gregor Wentzel, Karl Herzfeld and Paul Ewald. He also encountered Sommerfeld's assistant Wolfgang Pauli.

Sommerfeld's students were broadly educated in both classical and modern physics. They knew fluid mechanics and electromagnetic theory and were, of course, drawn up in the emerging field of quantum mechanics. Laporte's first paper on the diffraction of electromagnetic waves around a sphere was published just before his 21$^{st}$ birthday. This became his PhD thesis.

His work on spectroscopy started soon thereafter. While unraveling the spectra of vanadium and iron, Laporte formulated the selection rule that today bears his name[1], and which is an early manifestation of parity conservation.

Otto Laporte came to the US in 1924 as one of the first Rockefellar fellows. He joined University of Michigan as an instructor in 1926, where Harrison Randall was soon to attract several illustrious names in physics, including George Uhlenbeck and Samuel Goudsmit.

In 1944 Laporte began, in effect, a new career in fluid mechanics. His first paper established the formula for the lift of an airfoil of elliptical outline. Two years later he began experimental studies using a shock tube.

Laporte was a charter member of the APS Division of Fluid Dynamics and served as chair of the Division in 1965.

Laporte died of rapidly progressing cancer on March 28, 1971. His death came after his name had been slated for presentation for election to the National Academy of Sciences at its annual meeting in April of that year. Taking an action it had never taken before, the Academy elected Otto Laporte to membership, posthumously. (For additional biographical information on Otto Laporte see his obituary for the National Academy of Sciences[2].)

Upon his death, his friends instituted an annual Memorial Otto Laporte Lecture, given for the first time in 1972 by Richard G. Fowler. This lectureship was endowed and turned into a regular award of the American Physical Society in 1985.

The outline of my Otto Laporte lecture is as follows: First, I will define what is meant by chaotic advection. Next, I will highlight some works that one can see as precursors to chaotic advection, works that contained important aspects of the concept but didn't quite capture it or, somehow, didn't 'take' with the fluid mechanics community at that time. Third, I will outline how chaotic advection did become established in the early 1980's. Fourth, I will mention some areas of application and of current research that seem to me to hold particular promise for the future.

## 2. CHAOTIC ADVECTION

When a particle moves with the fluid, we speak of advection, sometimes *passive* advection to emphasize that the particle is so light and inert that it can do nothing but follow the fluid, instantaneously adjusting its own velocity to that of the ambient flow. We may write

$$\mathbf{V}_{\text{particle}} = \mathbf{V}_{\text{fluid}} \tag{2.1}$$

as the formal statement of passive advection. In particular, the kinematics of the fluid itself is such that each fluid particle undergoes passive advection.

The particle velocity, $\mathbf{V}_{\text{particle}}$, is, of course, given by the rate of change of its position:

$$\mathbf{V}_{\text{particle}} = \left(\frac{dx}{dt}, \frac{dy}{dt}, \frac{dz}{dt}\right), \tag{2.2}$$

where (x ,y, z) is the position vector of the particle, here written in ordinary Cartesian coordinates.

The fluid velocity, I shall assume, is given by other considerations, which involve the solution of some set of partial differential equations, such as the Euler equations or the Navier-Stokes equations or the Stokes equations. We assume that this is taken care of elsewhere – all that interests us is that we are, somehow, given the fluid velocity components u, v and w as functions of the coordinates and the time, i.e., we are provided with

$$\mathbf{V}_{\text{fluid}} = (u(x, y, z, t), v(x, y, z, t), w(x, y, z, t)). \tag{2.3}$$

The condition that particle velocity equals fluid velocity then leads to a system of ODEs that I call the *advection equations:*

$$\frac{dx}{dt} = u(x, y, z, t), \tag{2.4a}$$

$$\frac{dy}{dt} = v(x, y, z, t), \tag{2.4b}$$

$$\frac{dz}{dt} = w(x, y, z, t). \tag{2.4c}$$

You will find these in many introductory texts, for example, on page 71 of *Fundamentals of Hydro- and Aeromechanics* by Prandtl and Tietjens in a chapter entitled 'Methods of Description.' We use two 'methods of description' in fluid mechanics, the Lagrangian method, where individual particles are tracked, and the Eulerian method, where one uses fields rather than particles[3]. The advection equations very definitely belong to the Lagrangian description of fluid motion.

From the vantage point of dynamical systems theory, three ODEs, such as (2.4), are more than enough for producing non-integrable or chaotic dynamics. The right hand sides do not even have to be terribly complicated. Thus, in 1963 Lorenz showed that three equations with simple quadratic couplings could be non-integrable[4]. It is important to emphasize, however, that Lorenz worked with three ODEs in his model of thermal convection because he truncated a large, in principle infinite, system of such ODEs to three equations. In the advection problem no such truncation takes place. There are three equations because we are dealing with motion in 3D space. Non-integrable behavior is, in this sense, a rigorous property of flow kinematics.

Returning to (2.4) we see that in 3D the flow need not be time-dependent in order to have chaos. Steady flows will do. In 2D we do need time-dependent flow to produce chaotic particle motion. Steady, two-dimensional advection is integrable.

There is an important twist to all this for 2D, incompressible flow, because in that case the velocity is derived from a streamfunction, $\psi$, through the familiar formulae

$$u = \frac{\partial \psi}{\partial y} \quad ; \quad v = -\frac{\partial \psi}{\partial x}. \tag{2.5}$$

Combining these with the advection equations turns them into something we recognize from dynamics, namely Hamilton's canonical equations for a one-degree-of-freedom system:

$$\frac{dx}{dt} = \frac{\partial \psi}{\partial y} \; ; \quad \frac{dy}{dt} = -\frac{\partial \psi}{\partial x} . \tag{2.6}$$

The streamfunction plays the role of the Hamiltonian! The coordinates of the particle, x and y, are the conjugate variables. You can choose either one as generalized coordinate. The other cartesian coordinate then is the conjugate generalized momentum. (This is an extreme example of the symmetry between generalized coordinates and momenta in Hamilton's formulation of dynamics.) Phase space in this problem is configuration space.

In short, two-dimensional kinematics of advection by an incompressible flow is equivalent to the Hamiltonian dynamics of a one-degree-of-freedom system. This applies regardless of whether the dynamics of the fluid itself is viscous or inviscid. There is no contradiction or paradox here. The Hamiltonian nature of the kinematics derives just from incompressibility. This is independent of whether the fluid motion dynamically is dissipationless or not.

The notion that particle motion in a 2D incompressible flow can be viewed as a Hamiltonian dynamical system with one degree of freedom was, I believe, generally known in the fluid mechanics community for many years. However, it may have been considered a sterile, formal observation. The possibility that a time-dependent Hamiltonian system with one degree of freedom can be chaotic was surely understood by the mid-1960s. Nevertheless, the connection between these two ideas and, thus, the introduction of the term chaotic advection, was not realized until the early 1980s. I shall discuss the timeline of those developments shortly. Before doing so I want to highlight some earlier works that contained germs of the idea.

## 3. PRECURSORS

The first paper I wish to single out is by Carl Eckart, a contemporary of Laporte. His name is today associated with the Wigner-Eckart theorem in quantum mechanics, a sophisticated symmetry result and selection rule that, I suspect, includes Laporte's rule as a special case. In 1948, while working on geophysical fluid dynamics – Eckart served as the director of the Scripps Institution of Oceanography – he published a paper in *Journal of Marine Research*[5]. In it he makes a case for using the two words 'stirring' and 'mixing' to distinguish different physical processes. I quote from the paper: "...advection alone will ultimately increase the mean value of any initial gradient...," Eckart wrote. "This effect of advection is appropriately called stirring." And: "The effect of conduction or diffusion is to decrease the mean value of the gradient. This is appropriately called mixing... Ordinarily, the early stages of the process... will be dominated by the advective processes... These may so increase the mean gradient that the mixing process will ultimately dominate... Viscosity, if not counteracted by other factors, tends to stop the stirring ... before an appreciable amount of mixing can occur." I find these qualitative ideas and the terminology to be very insightful and useful, and I want to advocate Eckart's proposed use of the two words to distinguish the 'mechanical' and 'molecular' physical processes that produce mixing.

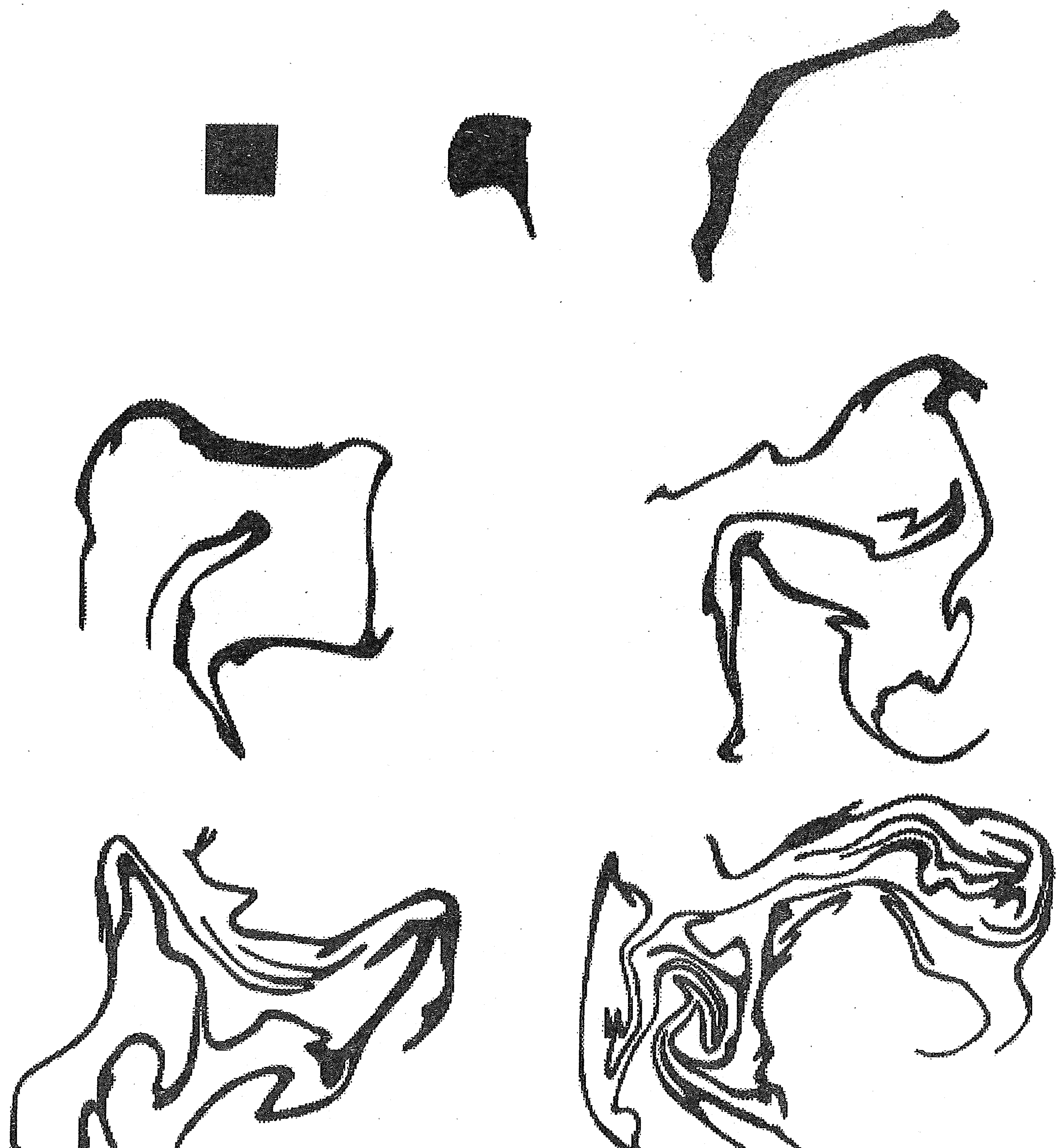


Figure 1: Stirring of a 'blob' of marked fluid by 2D, time-dependent, laminar flow (Welander, 1955)

The second precursor is a paper by Pierre Welander, a doctoral student of the Swedish physicist Oskar Klein, with whom he worked on kinetic theory. Fresh off his doctorate, Welander joined Rossby's institute. Using a gently stirred tank of fluid in essentially 2D motion, Welander introduced a blob of slowly diffusing dye into the flow and watched the changes in its spatial pattern. In his experimental pictures (Figure 1) one sees that the blob becomes highly ramified, with increasingly fine structure teased out into long arms. One might think the underlying flow was turbulent, but it wasn't. The instantaneous streamline pattern of the flow was just a system of nested ovals of, roughly speaking, the scale set by the outer boundary of the vessel.

So here is a paradox: How does a flow of a certain instantaneous scale produce patterns in an advected marker that are much, much smaller than the flow scale? How do these strange curls and tentacles in the pattern develop when the streamlines do not display them?

At the time Welander did not know about chaos (although a decade later he would become one of the pioneers of the subject) and so attempted to use ideas from equilibrium statistical mechanics, in particular the 'ergodic theorem', according to which a system would explore more and more of its phase space, eventually reaching it all. This was, somehow, the reason the advected patch spread out as if to cover every part of the flow domain.

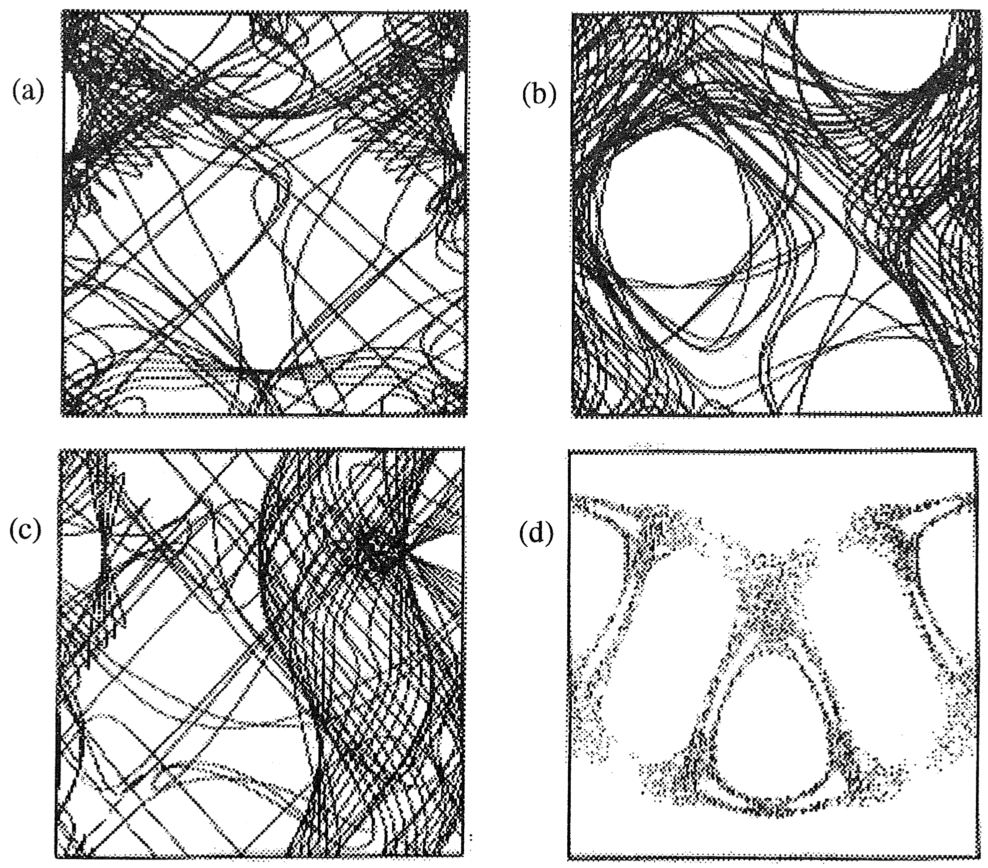



Figure 2: (a)-(c) Particle trajectories as viewed from three perpendicular directions in the so-called ABC flow given by (u,v,w) = (sinz + 0.65 cosy, sinx + cosz, 0.65 siny + cosx); (d) intersections of the trajectory shown in (a)-(c) with a plane parallel to one of the faces of the periodic cube. (Reproduced from H. Aref, S. W. Jones, S. Mofina & I. Zawadzki, "Vortices, kinematics and chaos." *Physica D* **37**, 423-440 (1989 with permission from Elsevier Science.)

The third precursor is a set of two, linked papers by V. I. Arnol'd and M. Hénon from the mid 1960's[7-8]. There is, of course, no doubt that these pioneers of dynamical systems theory understood exactly what chaos or integrability of the advection equations is all about.

Arnol'd reasoned[7] that in Beltrami flow, where velocity and vorticity are everywhere parallel, the Lamb vector, $\mathbf{V} \times \boldsymbol{\omega}$, the vector product of velocity and vorticity, which in steady, inviscid flow is the gradient of the Bernoulli integral, would vanish everywhere, and so would not constrain particle paths to two-dimensional sheets. Hence, as a manifestation of non-integrability of the advection equations, particles would wander throughout a subset of the flow volume.

In Figure 2 three perpendicular views of a particle path in a so-called ABC flow bear this out. If the path is cut by a plane through the cube, as in the fourth panel, the points of intersection fill a region of the plane, indicative of the wandering of the path in space. Hénon computed such surfaces of section, remarkable for the time, and verified non-integrability of the advection equations for these special flows[8].

Why didn't these studies 'take' in the fluid mechanics community? Was it the terse statement in French? Was it the somewhat contrived flows that were used? I don't know. Maybe it was thought that the phenomenon was limited to Beltrami flows. Whatever the reason, chaotic advection had to wait almost another 20 years.

In a paper by Berry *et al.*[9], published in 1979, the authors speculate on the shape of a curve subjected to a general, two-dimensional, area-preserving map, as would be the case for a material curve in periodic, 2D, incompressible flow. They distinguish the 'signatures' of elliptic and hyperbolic fixed points, referring to the wrapping-around action in the vicinity of the former as a 'whorl' and the stretching and compression in the tangle close to the latter as a 'tendril'. They go on to say that a continuous curve will "evolve into a fantastic shape incorporating both whorls and tendrils... Its curlings and flailings are reminiscent of cream spreading on coffee, and suggests that the study of generic area-preserving maps of curves on a plane, or surfaces in space, might be a profitable way to study turbulent mixing." Except for the word 'turbulent', which is unnecessary in the sense that laminar flows can accomplish this, this is of course exactly what will happen when a material line is advected chaotically. Again, this insight was a few years ahead of its time and seems to have left no impression at all on the fluid mechanics community. One should probably excuse fluid mechanicians for not noticing this paper – it was published in a mainstream physics journal, and its title was "Quantum maps"!

## 4. CHAOTIC ADVECTION INTRODUCED

I come now to the time-line for the introduction of the idea of chaotic advection in fluid mechanics. (On the transparency introducing this section of the talk, I had a snapshot of Laporte with Erwin Schrödinger, one of the founders of quantum mechanics, and Harrison Randall, the very energetic chairman of the Michigan Physics Department at the time.)

Many readers will have participated in the Geophysical Fluid Dynamics program at the Woods Hole Oceanographic Institution. I am a 1980 alumnus of that program. In 1982 a symposium on environmental tracers was held in conjunction with the Woods Hole GFD program, and I presented a paper there. The schedule shows that I spoke on Tuesday, July 27, at 2:00 p.m. The title of my talk was "An idealized model of stirring." I presented results on the 'blinking vortex model' described below. The abstract[10], published in the proceedings, concluded with this sentence: "Essentially what is being proposed is the existence of a new advective regime, intermediate between turbulent and laminar advection, which one might call 'chaotic advection'." So, this is, to the best of my knowledge, the first time the term 'chaotic advection' was used in the scientific literature.

A week or so later, when I submitted my abstract for the APS meeting that year, the term 'chaotic advection' had migrated to the title and my contribution at the Rutgers meeting was entitled "Stirring by chaotic advection." As usual, the abstract was published in the *Bulletin of the American Physical Society*[11].

This was also the title of the paper[12] subsequently submitted for publication to *Journal of Fluid Mechanics* on March 30, 1983. I note the date, because on April 20, 1983, just three weeks later, Wayne Arter sent in a paper on Rayleigh-Bénard convection to *Physics Letters*. Arter's paper[13] contains the idea of chaotic advection. In a sense, it is similar to Hénon's study[8] but uses a more realistic and more readily accessible flow. Arter wrote: "Stream-lines of steady Rayleigh-Bénard convection with square planiform are displayed using Poincaré maps. As the second order mode becomes more important the flow becomes ergodic from the boundaries inward, like a perturbed, integrable Hamiltonian system." And: "It seems experimenters in convection have been unwittingly studying a perturbed, integrable Hamiltonian system; e.g., there are obvious implications for the transient mixing of a scalar contaminant in a developed velocity field."

What does the 1984 paper, Ref.12, contain? First, there is a statement of the general idea of chaotic advection, as just outlined in §2. Then a simple model of what the chemical engineer would call a 'batch stirring device' is introduced. This model later became known as the 'blinking vortex model'. In the model the fluid to be stirred is confined to a circular disk. Incompressible, 2D, inviscid flow is assumed throughout. The agitators doing the mixing are represented by bound point vortices under the investigator's control in the sense that they can be turned 'on' and 'off' (or they could be moved about, but this was not pursued in the paper). An agitator is 'on' when the corresponding point in the flow domain acts like a point vortex. The flow in the disk is then the flow induced by this point vortex modulo the slip boundary condition on the bounding circle. When the agitator is 'turned off', it is assumed that the fluid flow stops immediately. The other agitator can now be turned 'on', and so forth. The model is obviously kinematic rather than dynamic.

The motivation for the model came from work on interacting point vortices done with Neil Pomphrey and published a few years before[14]. I cannot elaborate here, but readers who know that literature will quickly see the connection. Two interacting point vortices in a circle will produce chaotic advection, but the link to practical stirring devices seemed tenuous. Hence, the modification to externally controlled agitators.

The blinking vortex model was analyzed using standard techniques from the theory of dynamical systems, in particular the numerical construction of Poincaré sections for different values of the time, T, that each vortex-agitator was left 'on'. Since in this problem the flow plane coincides with phase space, as already noted following (2.6), a Poincaré section may be thought of as being generated by the stroboscopic illumination of advected points with the period of the stroboscope set to the switching time for the agitators. The numerically computed Poincaré sections clearly show the emergence of chaos in the advection. When the switching time is short, the flow sensed by a particle at some distance from the agitators is, roughly, like a steady flow. Indeed, the advection due to alternately pulsed agitators, in the limit of short pulses, is a crude algorithm for integrating the motion of a particle in the flow field due to both agitators being 'on' for all time! As the switching time becomes longer, more and more chaos is apparent in the computed Poincaré sections.

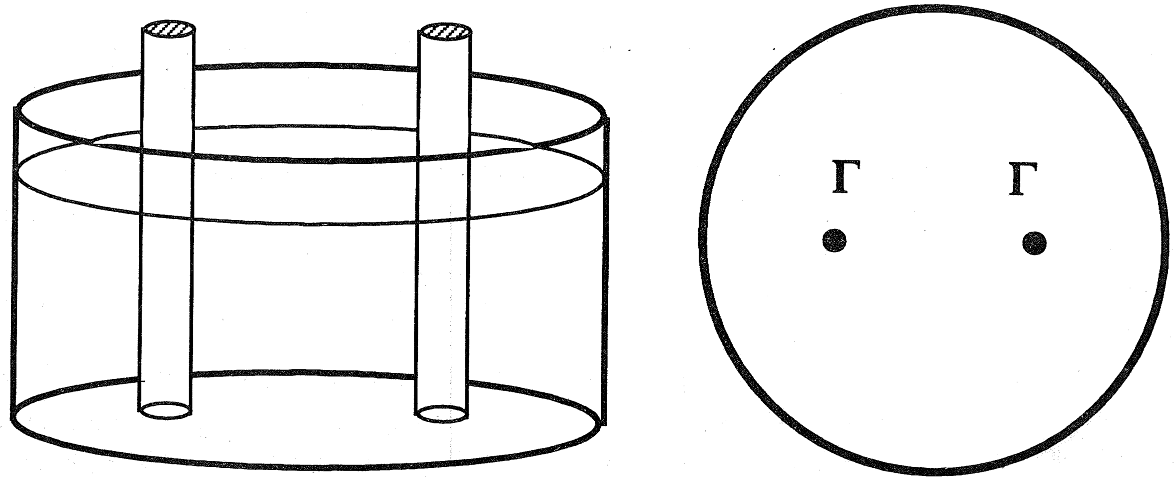



Figure 3: A 'batch stirring device' with two 'agitators' and its representation in the 'blinking vortex model' by a disk of fluid with two, bound, switchable point vortices.

To solidify the connection to a stirring process, numerical simulations were performed in which a blob of 10,000 points was passively advected by the blinking vortex flow. The computations clearly showed that the dispersion of the discrete particles during a fixed amount of time was much more dramatic when the switching time, T, corresponds to the chaotic regime than when it corresponds to the integrable or near-integrable regime. Since the flow is incompressible, the observed dispersion of particles signals tremendous stretching of material lines. Since the domain is bounded, this continual stretching implies repeated folding. There were other things in the paper, for example, a preliminary discussion of iterated maps and symmetries as applied to the blinking vortex flow problem. (The reader is referred to Ref.12 for details.)

How was the paper received? Well, here is the gist of the referees' reports obtained through JFM. The first referee wrote: "... a well written paper that gives a delightfully interesting example of chaotic advection. I think it will be a most useful addition to the literature." Obviously, this referee got it right. The paper did become a "most useful addition to the literature."

The second referee started his report thus: "This ... very neat but somewhat irritating paper... probably deserves publication although this reviewer has ... reservations regarding its relevance to the behavior or 'real' fluids (whatever they are).

The idea of interpreting particle paths as solutions of an ODE ... is nice, even if (as Aref points out) obvious. The few fluid mechanicians who have used the analogy fruitfully have been ignorant of recent developments in dynamical systems and hence have missed most of the benefits." He then made a variety of comments, none of them terribly difficult to deal with in a cosmetic revision of the text.

For workers familiar with dynamical systems theory the paper must indeed have been "neat but somewhat irritating." It was certainly true that similar ideas had been pursued for the motion of charged particles in electromagnetic fields, both in particle accelerators and in plasma devices. In those cases chaos was 'bad', something to be avoided. What is different in the fluid mechanics application is that, maybe for the first time, chaos is 'good'. If you want to stir efficiently, you want as much chaotic advection as you can get!

At the end of his report this referee concluded: "... this short paper has provoked me into a good deal of thought and long review. It surely should be published, after appropriate revision."

Then there was the third referee's report. I reproduce it verbatim: "I was not impressed by that paper since it seems to me that all the important points have been covered for general measure-preserving maps of the plane in MacKay's brilliant thesis, and that the special case of the flow induced by a moving vortex adds little of note or of hydrodynamical importance." Time has been less kind to this referee's assessment.

As a point of possible historical interest, I mention that two of the three referees would receive the Laporte Award in the years between 1984 and the present.

Fortunately we have editors and the paper was accepted. The editor's take on the above was that "the reports are generally favorable, except for the rather jaundiced reply of the [third] referee..." and that "... you may consider the paper to be accepted on a provisional basis, since I suspect that you will be able to satisfy the [second] referee." It took a fairly long time to get the manuscript into print, since I had to redo all the illustrations – the thousands of little dots depicting advected particles and points in Poincaré sections were, apparently, too small and the original figures did not reproduce well at Cambridge University Press. In the end all was resolved and the paper appeared as the lead article in volume 143 of *Journal of Fluid Mechanics,* the issue of June 1984.

The paper has remained popular, and the term 'chaotic advection' has stuck. (The paper was reproduced in 1987 in the book *Hamiltonian Dynamical Systems,* edited by R. S. Mackay and J. D. Meiss, published by Adam Hilger.) Figure 4 shows results obtained using the electronic version of the Science Citation Index. The solid bars show the number of citations of the 1984 paper as a function of calendar year. I am pretty confident of these results since I know most of the papers individually. The cross-hatched bars are supposed to give the number of articles that use the term 'chaotic advection' either in the paper title or in the abstract. I think the trend is correct but the absolute numbers seem somewhat low. Some authors use the term chaotic mixing, instead of chaotic advection. This is, in principle, undesirable given Eckart's very useful classification. Also, in the early years the term 'Lagrangian turbulence' had its followers but was soon deemed to be imprecise and inappropriate. Such variations in language can easily explain why the number of citations of the paper is larger than the number of uses of the exact term 'chaotic advection' in title or abstract.

On balance, I think it is safe to conclude that the concept and the terminology are here to stay. It has been thrilling to watch over the years this intellectual offspring take its first steps and gradually mature.

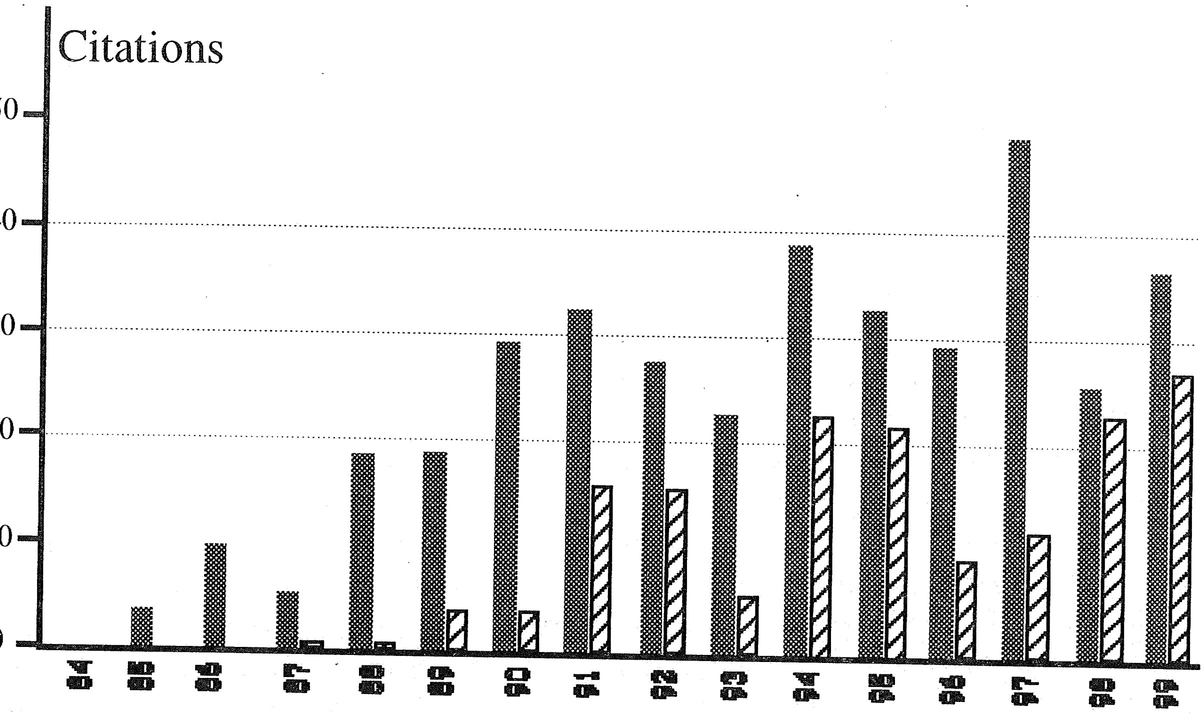



Figure 4: Solid bars: number of citations of *J. Fluid Mech.* **143** (1984) 1-23 as a function of calendar year; cross-hatched bars: number of articles that use the term 'chaotic advection' in either title or abstract as a function of calendar year.

## 5. FURTHER DEVELOPMENTS

It was immediately clear that the context in which to look for chaotic advection experimentally was viscous flows – in fact, Stokes flow, in which the entire flow field is prescribed by the motion of the boundaries, seemed ideally suited. So, the hunt was on for Stokes flows of sufficient complexity. The Stokes flow between rotating cylinders, one situated eccentrically within the other, was a perfect candidate. Balachandar and I, and unbeknownst to us, a group at Columbia University, quickly tooled up to do numerical simulations of chaotic advection in this flow.

The Columbia group, led by René Chevray and Michael Tabor, with PhD student Joel Chaiken, went further and set up an experiment, reported on in their 1986 paper[15] in the *Proceedings of the Royal Society.* The results, were very satisfying. The dye introduced into the flow distributed itself quite differently according to whether it was in a regular island or in the chaotic sea (the reader is encouraged to consult the plates and figures in their paper). Within islands one got a 'jelly-roll' structure, as one would expect from largely integrable advection. In the chaotic sea, however, the dye was entirely homogenized on the same time-scale, suggesting that much more diffusion had taken place due to rapid interfacial stretching. Chaiken *et al.* computed Poincaré sections for their flow, as Balachandar and I had done independently[16], and were able to correlate the computations completely with experimental observations. Indeed, on occasion computation suggested small islands that had been missed in an initial run of the experiment, but which were then found.

Thus, reference 15 provided the first experimental verification of chaotic advection in a time-dependent, 2D viscous flow in the sense that a detailed comparison was achieved, over an extended range of flow control parameters, between a computed Poincaré section and the corresponding flow visualization signature. It was important in those early days of the subject that the flow be known analytically so that secondary flows or other physical mechanisms could be ruled out as sources of the complex patterns observed. Knowing the flow analytically also allowed essentially perfect numerical control over the advection of the particles in computer simulations.

At the same time an experiment using a cavity flow with moving walls by Chien, Rising & Ottino[17] identified the presence of an embedded horseshoe map via flow visualization. This flow was not determined analytically at the time, so a direct comparison between experiment and numerical computation could not be performed. The identification of the horseshoe map was posited as direct, topological evidence of chaotic kinematics in the flow.

Many experiments now followed, and many very pretty illustrations of the phenomenon have been produced. For example, the cover illustration for the January 1989 issue of *Scientific American,* which appeared in conjunction with Ottino's article for that magazine, showed again the complex patterns produced by very low diffusivity dye in Stokes flow between eccentric rotating cylinders. The intricacy of chaos, usually confined to a rather inaccessible phase space, was here being displayed in real space, thanks to Eqs.(2.6). Many magazine editors and others involved with articulating science to the general public found these beautiful images to be quite irresistible.

It is impossible to follow the developments from here on in the same level of detail, since once the connection between advection and chaos in a dynamical system had been revealed, a multitude of applications suggested themselves, and the entire toolkit of dynamical systems was applied to the problem of flow kinematics. Thus, I will only mention a few highlights.

Ottino's book[18], part monograph, part graduate text, appeared in 1989, just a few years after the first research papers on the subject had been published. (It was published in the book series on applied mathematics that David G. Crighton – who won the Otto Laporte Award in 1998 – and I had started shortly before.) This book played an important role in 'spreading the word' about the role of chaotic dynamics in fluid mixing processes to various scientific and engineering communities. Also published that same year, Michael Tabor's text on nonlinear dynamics[19] included a section on chaotic advection.

The following year an IUTAM symposium on the *Fluid Mechanics of Stirring and Mixing* was held in San Diego. Chaotic advection was a major theme of the meeting. The proceedings[20] appeared as a special issue of *Physics of Fluids.*

The first major meeting to have the term 'chaotic advection' in the title was, I believe, the combined NATO and European Geophysical Society workshop on *Chaotic Advection, Tracer Dynamics and Turbulent Dispersion,* held in Italy in 1993. The proceedings[21] were published as a special issue of *Physica D.*

In the year 2000, it might be natural to gauge the popularity of the subject by entering 'chaotic advection' into the various search engines on the world wide web. I did that at the end of October 2000. Depending on the search engine, I obtained anywhere from 125 'hits' to more than 100,000. I suspect this says as much about chaotic advection as it does about the scope and accuracy of search engines!

Today 'chaotic advection' appears as one of the keywords that authors of papers in this journal may use for classifying their papers.

## 6. OUTLOOK

Chaotic advection has seen numerous applications in diverse areas of fluid mechanics[22] and the list of applications continues to grow. Some of the most important may be those addressing the stirring of fluids on geophysical or planetary scales. Numerical simulations of stirring due to convective motions in Earth's mantle show every sign of chaotic advection (in the sense of Fig.1), in the sense of folds and layers on smaller and smaller scales. This idea can be pursued down to the scale of the striations seen in individual rocks. The fluid agitation due to periodic tidal ebb and flow in shallow waters has suggested interpretation in terms of chaotic advection rather than the prevailing turbulence modeling[23]. A most interesting application has been to the flow dominated by an azimuthal jet, such as might be encountered in the polar night jet surrounding the ozone hole at Antarctica[24]. Chaotic advection, thus, may have a significant role to play in global climate change.

I conclude with three recent developments that seem to hold particular promise. I have singled out two new application areas in the engineering sciences – microfluidic devices and materials processing – and what I believe to be the most significant advance in the theory in several years: the use of ideas from topology to 'build in' chaotic advection in the design of a stirring device.

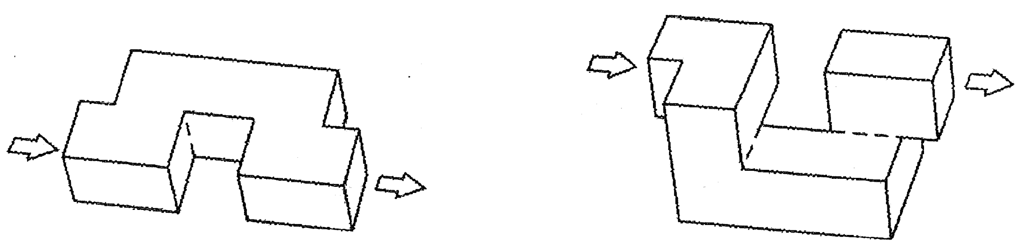


Figure 5: Geometry of the 'basic cell' used in the MEMS devices manufactured by the Illinois group.
Left: The 2D 'square wave' mixer. Right: The 3D 'serpentine' mixer.
(Reprinted from Ref. 27 with permission of Elsevier Science.)

Let us first talk about MEMS, in particular their application in biofluidics, where the objective is to perform within the micro-channels of a chip some of the feats usually accomplished with ordinary lab-scale equipment. Since we are talking about fluid motion in tubes that are the breadth of a human hair, the Reynolds number of the flow is small, certainly less than 100, and often of order 1. Although dimensions are small, diffusion times may be long compared to residence times in the device, so some kind of stirring is required. Furthermore, since the liquids are biofluids of various kinds, limiting the total strain on an embedded macromolecule is desirable. Chaotic advection provides an attractive engineering solution for achieving efficient stirring in this context.

The first biofluidics stirrer based on chaotic advection, built by Evans, Liepmann and Pisano[25], was modeled on the pulsed source-sink flow that Scott Jones and I had studied a decade earlier[26]. The device, unfortunately, has several moving parts that present problems for manufacturability and reliability of operation.

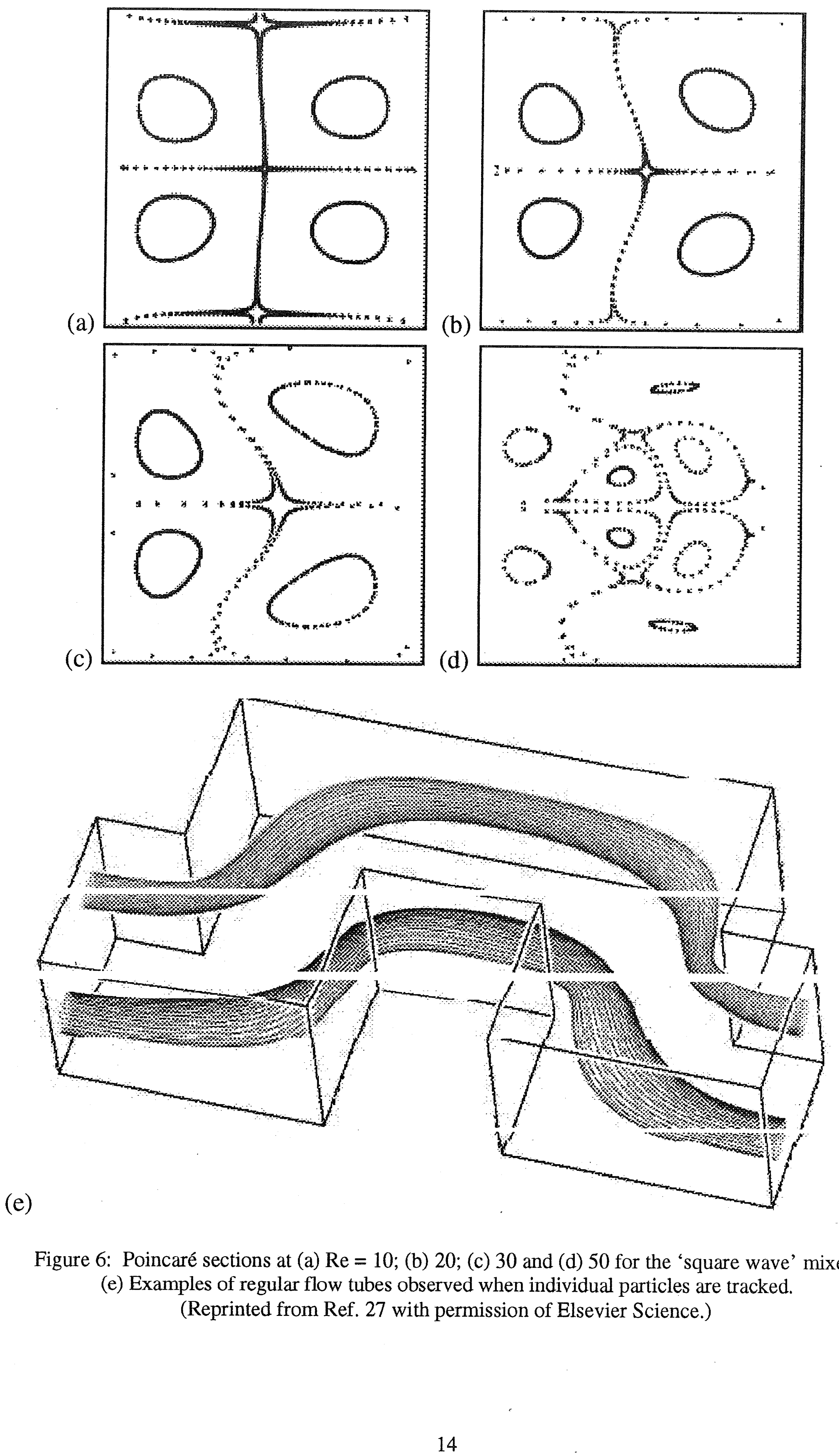



Figure 6: Poincaré sections at (a) Re = 10; (b) 20; (c) 30 and (d) 50 for the 'square wave' mixer; (e) Examples of regular flow tubes observed when individual particles are tracked. (Reprinted from Ref. 27 with permission of Elsevier Science.)

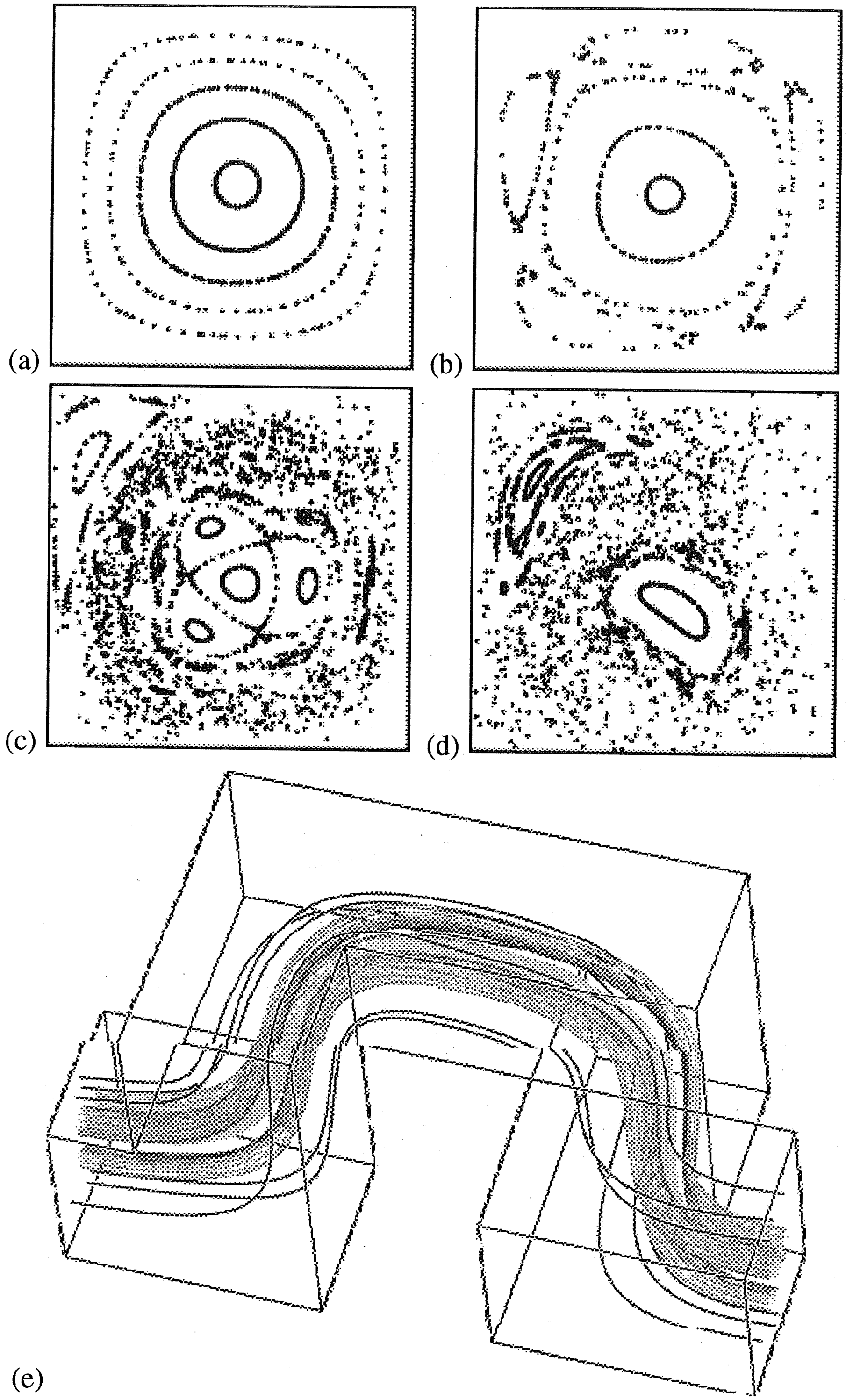



Figure 7: Poincaré sections at (a) Re = 1; (b) 5; (c) 10 and (d) 20 for the 'serpentine' mixer; (e) Examples of regular flow tubes and chaotic trajectories observed when individual particles are tracked. (Reprinted from Ref. 27 with permission of Elsevier Science.)

Static mixers are more promising. The devices produced by the Illinois group[27] (see Fig.5) use the twisted pipe flow, studied by Jones, Thomas and myself[28], as a guide. Flow progression

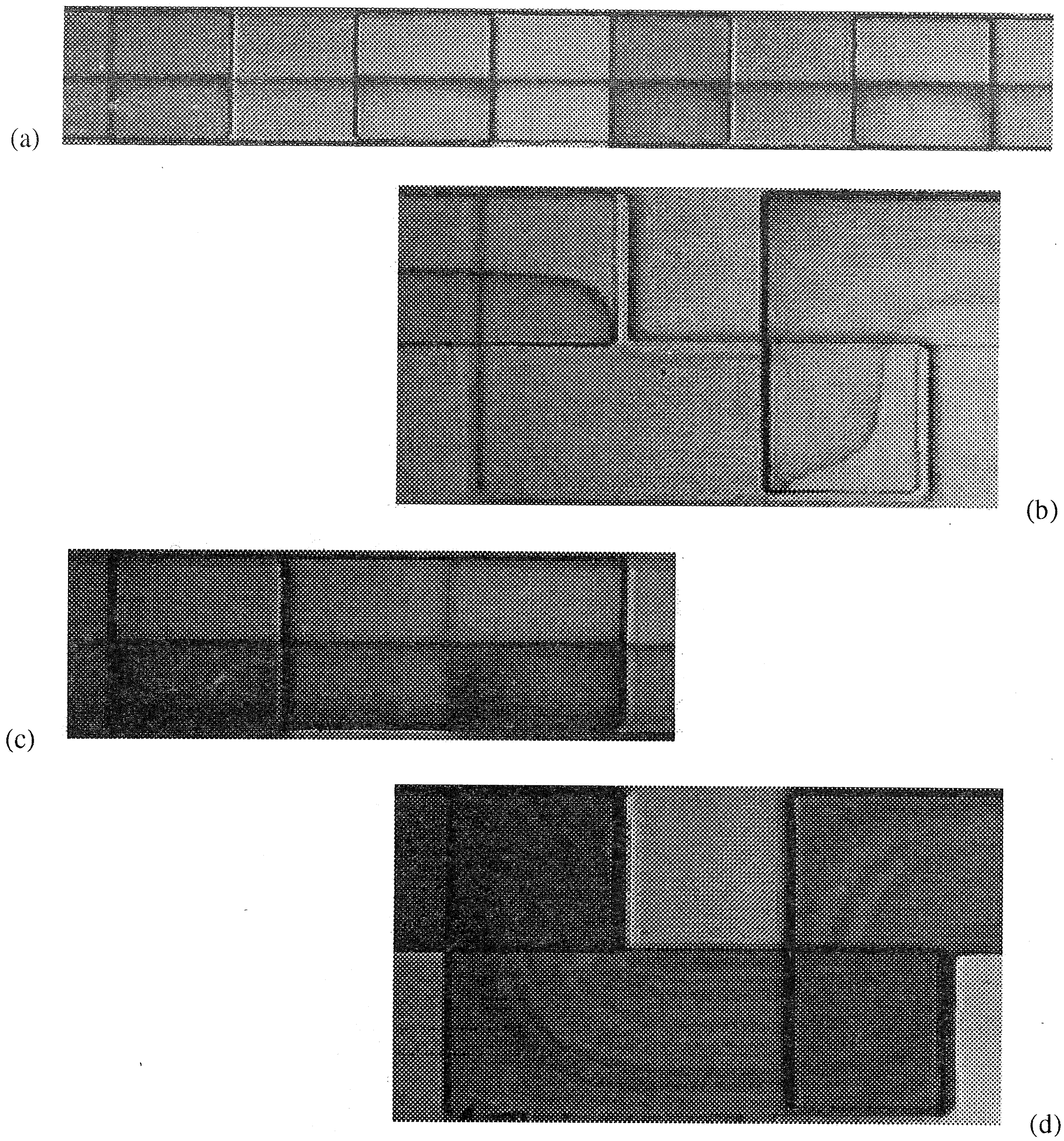
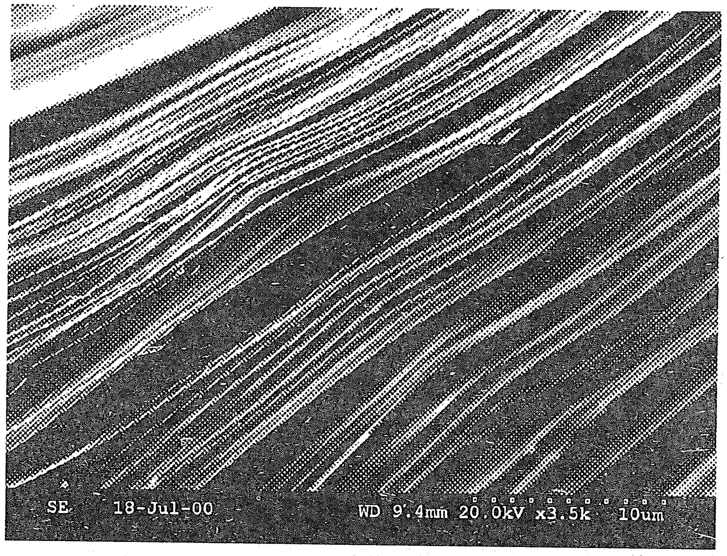
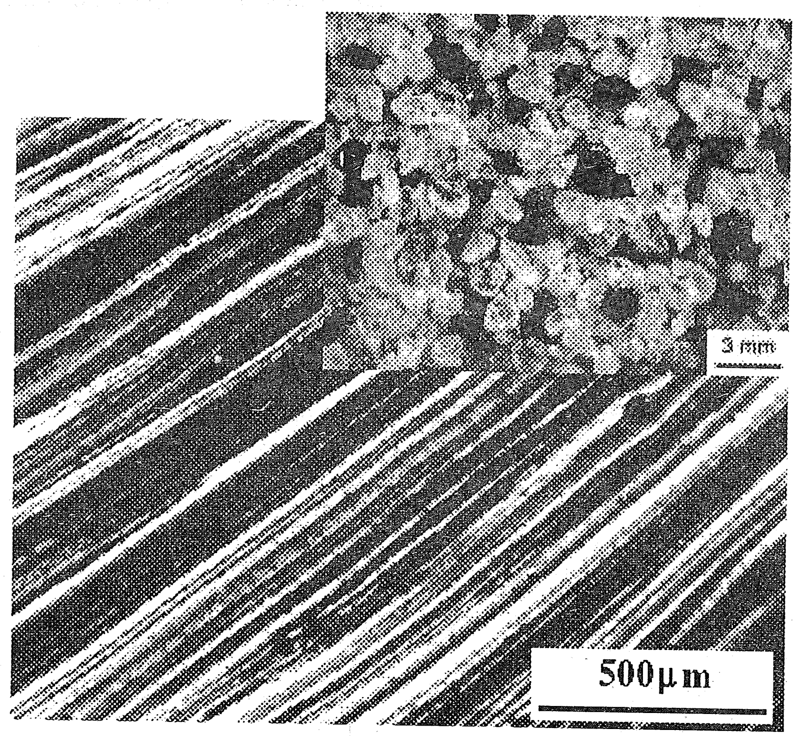



Figure 8: Flow visualization studies of fluid mixing in MEMS devices. The marked fluid results from an acid-base reaction between the fluid entering above and below the midline at the entry to the channel. (a) Flow in the 2D 'square wave' mixer, the first two segments viewed edge on at Re ≈ 10; (b) Comparison picture of the first segment of the 3D 'serpentine' mixer at Re ≈ 10; (c) Results for the 'square wave' at segment 14; (d) Comparison results for the 'serpentine' at segment 11. (Reprinted from Ref. 27 with permission of Elsevier Science.)

along the tube now substitutes for the role of time in the unsteady 2D flow models. At the small but finite Reynolds numbers in question a secondary flow pattern is produced in the cross-sectional plan. In the 'serpentine' geometry the twist from segment to segment rotates the secondary flow from segment to segment. To a particle flowing down the tube it appears as though the flow pattern in the cross-stream direction is being periodically re-oriented. True, the particles do not all feel the reorientation at the same times, since they propagate down the tube with different speeds. Nevertheless, the alternating secondary flow pattern has the expected and desired effect.




(a)




(b)

Figure 9: Materials processing by chaotic advection. (a) Multilayer films formed in an extruded 2.5 mm filament consisting of two thermoplastics (from Ref.31 with permission from Elsevier Science). (b) Thin parallel striations formed among carbon black particles in polystyrene (from Ref.32 with permission from Sage Publications, Ltd.).

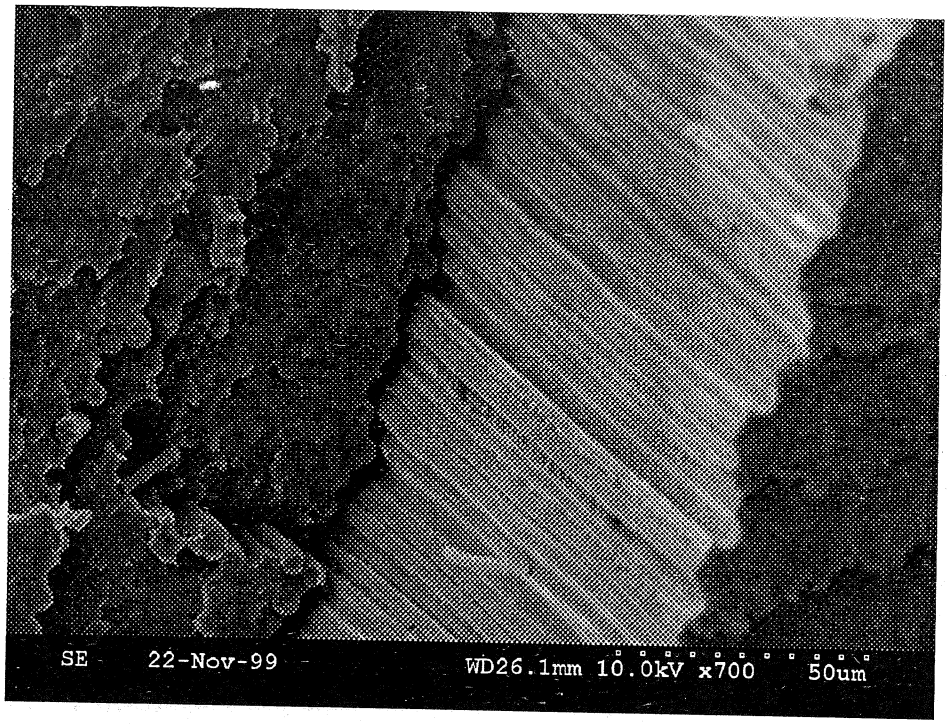
In place of Poincaré sections by stroboscopic illumination, as used in 2D time-dependent flow, we now construct sections by calculating the mapping of particles from one cross-sectional plane of the tube to the next, situated at a geometrically similar location along its length. This is, of course, an approximation since at finite Reynolds number there will, in general, be an influence from the flow in the $n^{th}$ segment on that in the $(n+1)^{st}$, but it is still a useful calculation to guide the design and implementation of the device.

In the MEMS application we built and compared two prototypes (Fig.5), a flat 'square wave' channel with 14 segments and a three-dimensional 'serpentine' channel with 11 segments. Calculations show[27] that in the square wave device the Poincaré section remains quite regular (Fig.6). If one tracks particles in this computed flow, one finds them to precess around in well-defined flow tubes. In the serpentine channel, on the other hand, calculations show a mixture of regular islands and chaotic regions in the section. Particle trajectories can be confined to islands but may also wander erratically across the cross section (Fig.7). This phenomenology is analogous to what one sees in the twisted pipe flow study of Ref.28. Cross-stream chaos is crudely similar to cross-stream diffusion and so the motion of the particles along the tube shares features with shear dispersion as discussed by G. I. Taylor in his seminal paper on fluid mixing[29].

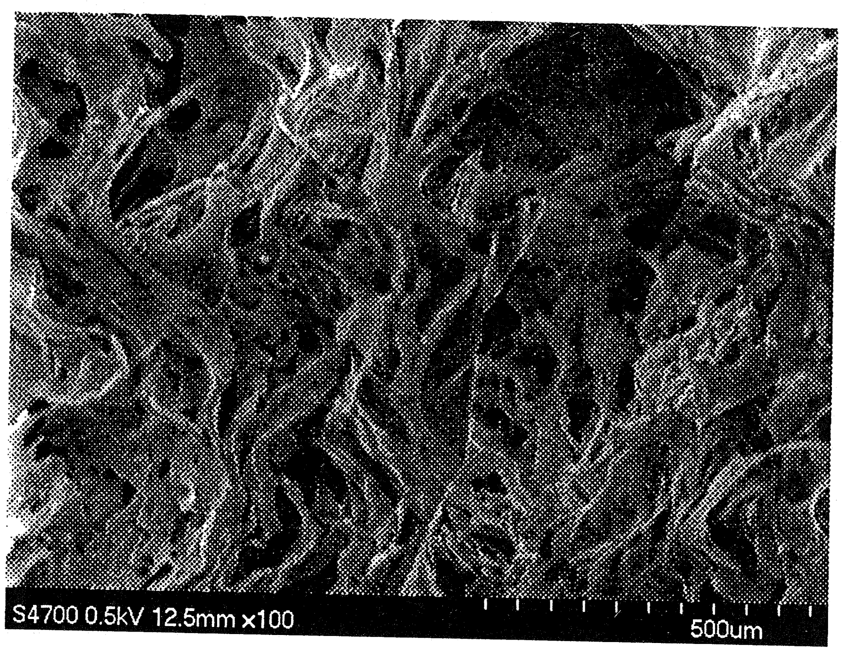
When flow visualizations are done, using an acid-base reaction, and the flow is viewed edge on, the results are quite dramatic (Fig.8). In the square wave channel one sees that almost half the cross-section remains clear even after several bends. In the serpentine channel, on the other hand, the colored product eventually fills the entire cross-section, indicating far superior mixing.

The next application I should like to mention is materials processing. The scales available due to chaotic advection reach from the size of the container to scales where the continuum hypothesis, on which the advection equations are based, breaks down or is cut off by physical effects that we have left out of the description, such as van der Waals forces. These consequences of chaotic advection have been exploited by David Zumbrunnen and his group at Clemson University. They are interested in the controllable formation of shapes in melts[30], including multi-layer films that are today largely produced by co-extrusion through dies. Co-extrusion provides films with fewer than 10 layers. Zumbrunnen's group injects polymer components and additives into a processing region where chaotic advection occurs. The components are stretched and folded recursively by the flow to yield sheets and multilayered films[31] (see Fig.9a). Particulate additives can also be organized into an abundance of filamentary chains so that plastics and glasses can be rendered electrically conducting at reduced percolation thresholds[32]. In order to get compositional uniformity, the authors began with a random mixture of polystyrene pellets and polystyrene pellets containing carbon black particles. The initial coarse mixture before melting and the resulting composite after chaotic advection are shown in Fig.9b. The striations render the composite electrically conducting. In conventional methods electrical conductivity is only obtained by chance at random locations and at higher additive concentrations, so the chaotic advection route to a conducting composite is very different and would appear to be superior.

These two examples involve primarily the 'stirring' phase (to use Eckart's classification). Later, interfacial tension, shear forces between components, and molecular forces (if internal length scales become sufficiently small) – the 'mixing' phase – lead to a variety of morphology changes. Orientation on molecular scales may even occur if components become confined to small regions spanning only nano-scales. The shape evolution is controllable by selecting the parameters in the chaotic advection, creating in effect an *in situ* processor. Rather than O(10) film layers as produced by conventional co-extrusion, Zumbrunnen and his group develop multilayer films consisting of hundreds or thousands of layers. In some cases, layer thicknesses less than 100 nm




(a)




(b)

Fig.10: Fibrous and interpenetrating morphologies due to film break-up in material processing using chaotic advection. (a) Fracture surface of an undrawn fiber with a fibrous microstructure consisting of 45% polypropylene by volume and polystyrene (from Ref.30 with permission of The Textile Institute). (b) SEM micrograph of interconnected low density polyethylene structures derived from simultaneous hole formation as chaotic advection proceeded in films formed in both the polyethylene and polystyrene components (from Ref.33 with permission of John Wiley & Sons.)

result. These are desirable because they are less prone to delamination, which is a common problem, and they may be better barriers to diffusing compounds. When the films break up upon continued stirring, novel fibrous[30] and interpenetrating morphologies[33] observed over large compositional ranges (Fig.10). Such morphologies provide tougher films due to the interconnectedness of the layers, and they may even lead to new electrically conducting materials.

Finally, let me say a few words about the application of ideas from topology to chaotic advection. The Stokes flow studies use configurations in which one or two cylinders are rotated in place to stir the fluid. Everyday stirring, of course, isn't done that way. When you stir your coffee, you don't usually put the spoon down and spin it in place! Rather you move it about in a circular or figure-eight like motion.

When this kind of stirring is considered, interesting topological considerations arise. For one stirrer and even for two stirrers things are relatively simple. But for three stirrers, there are two intrinsically different ways in which one can move them about, one that is topologically trivial, and one that it topologically profound. Little girls whose hair needs to be braided know all about this distinction.

To do the work justice would largely reiterate our paper cited as Ref.34. The application hinges on a beautiful piece of modern mathematics ('modern' by the usual standards of fluid mechanics!) due to Thurston with early contributions from the Danish mathematician Jakob Nielsen. This work gives a classification of the nature of the mapping of the disk of fluid when subjected to various stirring protocols. It is a topological theory, and so uses only the the idea of continuity of the fluid motion. The theory applies regardless of the underlying dynamics.

A simple experiment[34] shows the outcomes of two sequences using three stirring rods, and suggests profound differences in the outcome depending on the topology of stirrer motion. We hope we have found, in effect, a way to 'build-in' chaotic advection on the scale of the stirrer motion without the need to tune parameters after the fact. The application of Thurston-Nielsen theory to chaotic advection is, in my view, a very important and very interesting advance, and may in due course have important technological consequences.

I would like to think that such connections between deep insights into mathematics and applications to the physical world would also have pleased Otto Laporte. Laporte's parity rule, of course, has to do with the topological property of handedness. In looking over his publication list[2], I found that Laporte had published a paper on "Kowalewski's top in quantum mechanics," and another on Kepler ellipses. The issue of integrability versus chaos, and the nature of particle orbits would surely have interested him.

This is my account of the development of chaotic advection, an idea with important precursors due to some very deep thinkers in science. A very practical problem in fluid mechanics has been enriched by fundamental ideas from physics and mathematics, and through that enrichment now fosters advances in our technological capability, from MEMS to materials processing. Commercial devices utilizing chaotic advection are being built and patents embodying this mechanism have been issued. This interplay between the 'rational' and the 'practical', to use Newton's words, is what our science is all about. Applications of chaotic advection today span the range of length-scales from planetary atmospheres (scales of order 1,000 km or $10^6$m) to the materials processing of layers as thin as 100nm ($10^{-7}$m), i.e., more than a dozen orders of magnitude. The time scales involved range from seconds (for laboratory experiments) to geological time-scales (e.g., stirring of Earth's mantle). There is no reason that either or both of these ranges should not be extended in the future.

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