Advection of Active Particles in Open Chaotic Flows

Zoltán Toroczkai,[1,2] György Károlyi,[2,3] Áron Péntek,[4] Tamás Tél,[2] and Celso Grebogi[5,*]

[1]*Center for Stochastic Processes and Department of Physics, Virginia Polytechnic Institute, Blacksburg, Virginia 24061-0435*
[2]*Institute for Theoretical Physics, Eötvös University, Puskin utca 5-7, H-1088 Budapest, Hungary*
[3]*Research Group for Computational Mechanics, Technical University Budapest, Müegyetem rkp. 3, H-1521, Hungary*
[4]*Institute for Pure and Applied Physical Sciences, University of California at San Diego, La Jolla, California 92093-0360*
[5]*Institut für Theoretische Physik und Astrophysik, Universität Potsdam PF 601553, D-14415 Potsdam, Germany*
(Received 15 July 1997)

We investigate the evolution of active particle ensembles in open chaotic flows. The active processes of the type $A + B \rightarrow 2B$ and $A + B \rightarrow 2C$ are considered in the limit of weak diffusion. As an illustrative advection dynamics, we choose a model of the von Kármán vortex street, and show that the backbone of the active processes is the fractal structure associated with the passive dynamics' chaotic saddle. This fractal dynamics leads to singularly enhanced concentrations, resulting in a distribution of products that differs entirely from the one in conventional active processes. This may account for the observed filamental intensification of activity in environmental flows. [S0031-9007(97)05034-5]

PACS numbers: 05.45.+b, 47.52.+j, 47.70.Fw

There has been recent interest in the advection of active particles in chaotic hydrodynamical flows [1]. By active particles, we mean tracers that undergo certain changes due to, say, chemical or biological interactions, but do not modify the underlying flow. By chaotic, we mean time-dependent, nonturbulent velocity fields with chaotic tracer dynamics (Lagrangian chaos) [2]. The motivation is to understand the new features of chemical or biological processes induced by the underlying chaotic dynamics, and their implications to environmental flows, such as ozone reactions [3] or population of microorganisms in the sea [4].

In contrast to previous investigations restricted to flows in closed regions, our aim is to consider a class of two-dimensional open flows in which the velocity field in the far upstream and downstream regions is uniform but nonstationary in a bounded region. A well-known laboratory example is the flow around a cylinder. Similar situations can be observed in environmental flows, such as the fluid motion in the wake of a pillar or the motion of air behind an isolated mountain. A unique feature of such open flows with asymptotic simplicity is the pronounced and stable fractal character associated with the chaotic tracer dynamics [5–8], which is clearly measurable in experiments [9]. This tracer dynamics is governed by a nonattracting chaotic saddle [10] containing an infinite number of periodic and nonperiodic bounded tracer orbits which never reach the far upstream or downstream regions. These regions are, however, foliated by the saddle's stable and unstable manifolds, respectively. Although both the saddle and its manifolds are not space-filling fractal objects, the unstable manifold is the avenue of long-time propagation and transport of tracers in such flows [5–9].

The activity of the advected particles is assumed to be a kind of "infection" leading to a change of properties if they come close enough to each other. Particles with new properties are the products. We argue and show in this Letter that, in the limit of weak diffusion, there is an *enhancement* of activity around the chaotic saddle and its unstable manifold since it is there where the active tracers spend the longest time close to each other. Then, as the products are passively advected, they trace out the unstable manifold. (The enhancement of activity is meant, in comparison with nonchaotic, e.g., stationary, flows.) Characteristic quantities associated with this process are the escape rate κ from the saddle and the unstable manifold's fractal dimension D_0.

In this Letter, we also show that the distribution of products follows a *nontrivial dynamics* that can be derived from the properties of the passive advection and of the interaction rule of nearby particles. In particular, we point out that a steady state of finite productivity sets in after a sufficiently long time. Moreover, the newly born active tracers cover the branches of the unstable manifold approximately uniformly with an average width ε^* in the steady state. This implies that, on linear scales smaller than ε^*, fractality is washed out, but a clear fractal scaling of the material can be observed on larger scales with the *same* fractal dimension D_0 as that of the unstable manifold in the passive flow. As a consequence, the differential equation derived here for active tracer ensembles has the fundamental and novel feature of containing *irrational* D_0-dependent powers of the number of product particles, signaling a *singular* enhancement of productivity. This singularly enhanced rate of activity has profound practical consequences. It may account for the observed filamental patterns of intense activity in environmental flows [3], an effect that cannot be explained if one considers diffusion processes alone.

To fix a frame of mind for our discussion and analysis, we consider the activity in the flow to be of chemical origin. To be specific, we consider simple *kinetic* models [1] where two passively advected particles of different types undergo a reaction if and only if they come within a

 0031-9007/98/80(3)/500(4)$15.00

distance σ, which is the interaction range, and can also be considered as a *diffusion distance*. We study both (i) an autocatalytic process $A + B \rightarrow 2B$, and (ii) a collisional reaction $A + B \rightarrow 2C$. Component A will be considered as the background material covering the majority of the entire fluid surface. For computational simplicity we assume that reactions are instantaneous and take place at integer multiples of a time lag τ. An important parameter is $\nu = \kappa\tau$, the number of reaction events occurring on the characteristic time of chaos. The case of continuous time reaction is obtained in the limit $\tau \rightarrow 0$ ($\nu \rightarrow 0$) and $\sigma \rightarrow 0$. For generality, we also investigate cases where ν is of order unity which is of relevance for biological processes accompanying advection [11].

The flow chosen to illustrate these phenomena is the von Kármán vortex street in the wake of a cylinder [5–9]. The radius R of the cylinder and the period T of the flow are taken as the length and time units. For simplicity, we use an analytic model for the stream function introduced in Ref. [6]. This model has been motivated by direct numerical simulations at Reynolds number 250, and has been used successfully to reproduce qualitative features of the tracer dynamics. The escape rate of particles in the reaction free flow is $\kappa = 0.36$ and the fractal dimension of the unstable manifold is $D_0 = 1.61$, while the background flow velocity is $v_0 = 14$ [6].

In order to simplify *numerical* calculations, we project the tracer dynamics on a uniform rectangular grid of lattice size $\varepsilon_0 \leq \sigma$, covering both the incoming flow and the mixing region in the wake of the cylinder. If there is a tracer inside a cell, it is considered to be in its center. With this projection the tracer dynamics defines a mapping among the cells. If a tracer starting from the center of a cell is advected into another after time τ, then the latter cell is considered to be the image of the first one with respect to the dynamics [12]. One iteration of the mapping consists of advection followed by an instantaneous chemical reaction on this grid of cells. The simulation entails a repeated application of these steps.

(i) For *autocatalytic reactions,* a cell is considered to be occupied by reagent B if it is an image of at least one B cell. Otherwise, the cell is considered to contain A. In addition, if a cell contains B at the time of the reaction, all of the eight neighboring cells are infected by B. Consequently, ε_0 plays the role of the interaction range σ in our simulation. Initially, we introduce a seed of reagent B, typically upstream from the cylinder. Figures 1(a) and 1(b) display the spreading of B (black) in the course of time. Note the rapid increase of the B area and the formation of a filamental structure. The inset in Fig. 1(a) shows the number of B particles in the computational domain as a function of time. After approximately four periods, the chemical reaction takes the period of the flow and reaches a steady state. While in Fig. 1(b) the B distribution has a rather scanty appearance, right after the autocatalytic reaction [as shown in Fig. 1(c)], most of the filaments of the

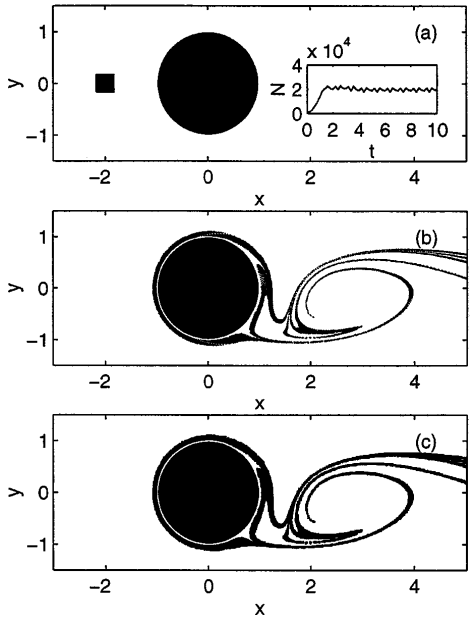

FIG. 1. Autocatalytic reaction: Time evolution of a square shaped droplet of B particles (black) in the background of A particles (white) for $\tau = 0.2$ ($\nu = 0.072$), $\varepsilon_0 = 0.01$, and at times (a) 0, (b) 2 (before the reaction), and (c) 2 (after the reaction). Inset in (a) shows the number N of B particles as a function of time.

manifold look wider. The two pictures correspond to two different coverages of the fractal manifold. The sudden increase of the coverage width at discrete times is due to our modeling of the chemical reaction as a "kicked" process. In the steady state, the reaction products are apparently distributed in strips of finite width along this manifold, and the B particles trace out a stationary pattern on a stroboscopic map taken with the period $T \equiv 1$ of the flow. On linear scales larger than the average width ε^*, the B distribution is a fractal. Figure 2 shows how the area of the black regions, right before the reaction, depends on the grid size $\varepsilon_0 = \sigma$. We find a scaling behavior with exponent 1.64, which is consistent with the unstable manifold's fractal dimension $D_0 = 1.61$ (cf. Fig. 2).

(ii) For *collisional reactions,* the cells are iterated *backward* in time, and the occupation of the neighboring cells is monitored. If, among the neighbors of the preimage, there exists a cell with either A or B, differing from the content of the preimage cell, then the original cell becomes C, just as if the preimage cell itself contains C. Initially, the flow consists of material A, into which we introduce B continuously along a line segment perpendicular to the background flow in front of the cylinder. Figure 3 displays a typical snapshot of the surroundings of the cylinder. Strips of product C separate the areas occupied by materials A and B. These strips are pulled along the unstable manifold of the chaotic saddle behind the cylinder. This implies again that the reaction essentially takes place along this manifold. After a short transient, a saturation

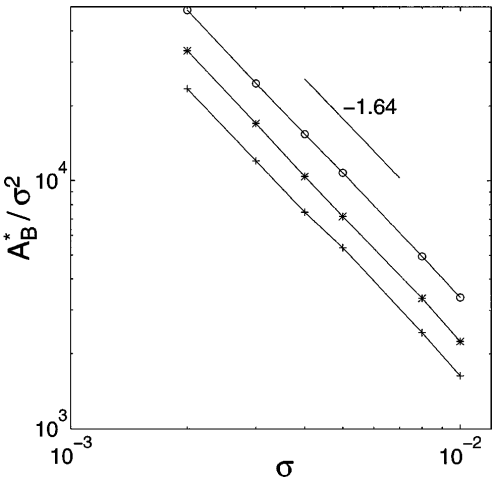

FIG. 2. The area occupied by B in the steady state scales as $\mathcal{A}_B^* \sim \sigma^{2-D_0}$ at different values of the time lag τ ($\circ$, $\tau = 1$; $*$, $\tau = 2$; $+$, $\tau = 3$) with $D_0 = 1.64$, which is in good agreement with the fractal dimension of the unstable manifold of the reaction free flow. Several runs were carried out with different $\sigma = \varepsilon_0$.

has been observed in the number of B or C particles on the stroboscopic map.

Next, we present a *theory*, valid for not too large time lags, which supports these observations. For simplicity, we treat the autocatalytic reaction $A + B \to 2B$ only. Let $\mathcal{A}_B^{(n)}(t)$ denote the area occupied by B inside the mixing region. Here $t \in [0, \tau]$ is the time after the nth reaction, and the total physical time is $n\tau + t$. During the time interval τ, only advection takes place, and $\mathcal{A}_B^{(n)}(t)$ decreases with the escape rate κ of the chaotic saddle. If the material is distributed along sufficiently narrow strips around the unstable manifold at $t = 0$, $d\mathcal{A}_B^{(n)}(t)/dt$ is proportional to $\kappa \mathcal{A}_B^{(n)}(t)$, and the area occupied by B at the end of the interval τ is $\mathcal{A}_B^{(n)}(\tau) = \mathcal{A}_B^{(n)}(0)e^{-\nu}$. At this time instant, a sudden reaction takes place, and the area of B becomes

$$\mathcal{A}_B^{(n+1)}(0) = \mathcal{A}_B^{(n)}(\tau) + R^{(n+1)}, \qquad (1)$$

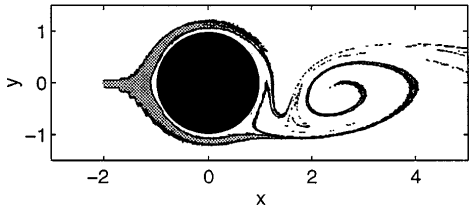

FIG. 3. Collisional reaction at time $n = 2$ (before the reaction). Initially the flow consisted of material A, into which reagent B was introduced along a line segment $x = -2$, $-0.05 < y < 0.05$ ($\sigma = \varepsilon_0 = 0.01$, $\tau = 0.05$). White denotes the background A, grey is B, while black is the product C.

where $R^{(n+1)}$ is the amount of reagent B appearing in the $(n + 1)$st reaction. The reaction takes place along the borderline between materials A and B. Let $\mathcal{L}$ denote the length of this borderline. The newly born B territory is then of area $\mathcal{L}\sigma$. A sufficiently long time after the onset of reaction the area of B is pulled into strips of more or less constant widths along the unstable manifold. Let $\varepsilon^{(n)}(t)$ denote the average width of the strips. By covering the full area occupied by B at any time instant by squares of linear size ε, the number of boxes needed for this coverage behaves as $N(\varepsilon) = \mathcal{H}\varepsilon^{-D_0}$ for $\varepsilon \geq \varepsilon^{(n)}(t)$. Here, $\mathcal{H}$ is the *Hausdorff area* of the manifold of dimension D_0. The area of B is then $\mathcal{A}_B = \mathcal{H}\varepsilon^{2-D_0}$. Since the number of boxes covering the perimeter is proportional to $N(\varepsilon)$ due to the filamentary structure, the length of the borderline separating A and B is, at the same instant of time, $\mathcal{L} = c\mathcal{H}\varepsilon^{1-D_0}$. Here, c is a number incorporating information about the *shape* of the fractal, a quantity which might depend on n but, if so, it is periodic with the period of the flow. By eliminating ε, we obtain that the perimeter length and the mass are related to each other via a power law: $\mathcal{L} = c\mathcal{H}^{1/(2-D_0)}\mathcal{A}_B^{-\beta}$ with $\beta = (D_0 - 1)/(2 - D_0)$. Since the manifold's dimension lies between 1 and 2, the exponent β is positive. The reaction term in (1) is then

$$R^{(n+1)} = \sigma\mathcal{L} = \sigma c\mathcal{H}^{1/(2-D_0)}\mathcal{A}_B^{(n)}(\tau)^{-\beta}. \qquad (2)$$

Thus we conclude that the reaction term contains a *negative* power of the area occupied by B. The less B material present, the more effective the reaction is, because the resolved perimeter is the larger. This *singular* behavior is due entirely to the fractality of the manifold, and it disappears for smooth manifolds (nonchaotic flows) when $D_0 = 1$. Therefore, the reaction equation (1) becomes

$$\mathcal{A}_B^{(n+1)} = \mathcal{A}_B^{(n)}e^{-\nu} + \sigma c e^{-\nu}\mathcal{A}_B^{(n)-\beta}, \qquad (3)$$

where we have used the rescaling $\mathcal{A}_B^{(n)}(\tau)/\mathcal{H} \to \mathcal{A}_B^{(n)}$ to eliminate the dependence on the Hausdorff area. This is a recursion (a discrete dynamics) for $\mathcal{A}_B^{(n)}$, expressing the amount of B before the $(n + 2)$nd reaction in terms of the amount before the $(n + 1)$st reaction. Since the map is one dimensional and strongly dissipative, it must possess attractors, in spite of the Hamiltonian particle dynamics. The advection by the flow (first term) and the reactivity (second term) are in permanent competition.

In steady states of the active process the two competitive terms balance each other. If c is constant, a fixed point $\mathcal{A}_B^*$ is found for the area occupied by B. It is easy to check that this fixed point is the only attractor. In the more general case when c is periodic with the period of the flow, a limit cycle is the attractor. In other words, the active process becomes *synchronized* to the underlying flow. The area occupied by B in the steady state turns

out to be proportional to $\sigma^{(2-D_0)}$, in accordance with the numerical observations (cf. Fig. 2).

The time-continuous case can be obtained in the limit $\tau \to 0$, $\sigma \to 0$ by keeping σ/τ constant. The discrete time mapping (3) becomes a differential equation: $\dot{\mathcal{A}}_B = -\kappa \mathcal{A}_B + c v_r \mathcal{A}_B^{-\beta}$. Here $\mathcal{A}_B$ denotes $\mathcal{A}_B^{(t/\tau)}$, and $\sigma/\tau \to v_r$ represents the *velocity* of the reaction front. For $D_0 = 1$ the differential equation describes a classical surface reaction along a line with front velocity v_r in the presence of escape. For $1 < D_0 < 2$ it represents a novel form of reaction equations containing also a negative power of concentration due to the fractality of the unstable manifold. Such processes are generalizations of classical surface reactions. [13]. In order to observe fractal patterns, the average covering $\varepsilon^* = c\sigma/\nu$ must be much less than unity. This implies $v_r \ll \kappa$, i.e., a reaction being *slow* on the characteristic scales of advection.

In conclusion, we have shown that active processes among advected particles lead to nontrivial distributions of products in open flows with asymptotic simplicity. The unstable manifold of the passive dynamics' chaotic saddle is the backbone for such processes. The product particles (and other components) accumulate along this fractal set as they fatten it up. (The inclusion of a weak molecular diffusion, leading to a random walk superimposed to the deterministic advection, would result in a further slight fattening up, in a renormalization of σ and ε^*.) As a consequence, the distribution of products is singularly enhanced, i.e., *the fractal manifold acts as a catalyst for the active process*. The latter is governed by a reaction equation different from conventional ones and contains a singular dependence on the concentrations due to the chaotic and fractal properties of the flow. The competition between advection and production typically leads to a steady state distribution synchronized to the flow's temporal behavior. These features are independent of the particular models used. In view of the fact that chaotic advection is generic, our findings should be valid for general active processes, including realistic chemical and biological reactions in environmental flows. They might also shed new light on the coupling between population or chemical dynamics and imperfectly mixed hydrodynamical flows [11].

Useful discussions with M. Kremliovsky, H. Lustfeld, Z. Neufeld, K. G. Szabó, and J. A. Yorke are acknowledged. Á. P. and Z. T. thank J. Kadtke, R. K. P. Zia and B. Schmittmann for their support and encouragement. This research has been supported by the NSF through the Division of Materials Research, by the DOE, by the US–Hungarian Science and Technology Joint Fund under Projects No. 286 and No. 501, and by the Hungarian Science Foundation T17493, T19483.

*Permanent address: Institute for Plasma Research, University of Maryland, College Park, MD 20742.

[1] G. Metcalfe and J. M. Ottino, Phys. Rev. Lett. **72**, 2875 (1994); Chaos Solitons Fractals **6**, 425 (1995).

[2] J. M. Ottino, *The Kinematics of Mixing: Stretching, Chaos and Transport* (Cambridge University Press, Cambridge, England, 1989); S. C. Jana, G. Metcalfe, and J. M. Ottino, J. Fluid Mech. **269**, 199 (1994); A. Crisanti *et al.*, Riv. Nuovo Cimento **14**, 1 (1991); Special issue of Chaos Solitons Fractals, edited by H. Aref, **4**, No. 6 (1994).

[3] S. Edouard *et al.*, Nature (London) **384**, 444 (1996); S. Edouard, B. Legras, and V. Zeitlin, J. Geophys. Res. **101**, 16 771 (1996); D. G. H. Tan *et al.* (to be published).

[4] C. H. Gibson and W. H. Thomas, J. Geophys. Res. **100**, 24 841 (1995); W. H. Thomas, C. T. Tynan, and C. H. Gibson, (to be published).

[5] V. Rom-Kedar, A. Leonard, and S. Wiggins, J. Fluid. Mech. **214**, 347 (1990); K. Shariff, T. H. Pulliam, and J. M. Ottino, Lect. Appl. Math. **28**, 613 (1991).

[6] C. Jung and E. Ziemniak, J. Phys. A **25**, 3929 (1992); C. Jung, T. Tél, and E. Ziemniak, Chaos **3**, 555 (1993); E. Ziemniak, C. Jung, and T. Tél, Physica (Amsterdam) **76D**, 123 (1994).

[7] D. Beigie, A. Leonard, and S. Wiggins, Chaos Solitons Fractals **4**, 749 (1994).

[8] Á. Péntek, T. Tél, and Z. Toroczkai, J. Phys. A **28**, 2191 (1995); Fractals **3**, 33 (1995); Á. Péntek *et al.*, Phys. Rev. E **51**, 4076 (1995); J. Kennedy and J. A. Yorke (to be published); M. A. Sanjuan *et al.*, Chaos **7**, 125 (1997); Phys. Rev. Lett. **78**, 1892 (1997); Z. Toroczkai *et al.*, Physica (Amsterdam) **239A**, 235 (1997).

[9] J. C. Sommerer, H.-C. Ku, and H. E. Gilreath, Phys. Rev. Lett. **77**, 5055 (1996).

[10] T. Tél, in *Directions in Chaos*, edited by Hao Bai-Lin, (World Scientific, Singapore, 1990), Vol. 3, pp. 149–221.

[11] G. Károlyi *et al.* (to be published).

[12] Note, that this is not a one-to-one mapping. Some cells may have the same image cell.

[13] L. D. Landau and E. M. Lifschitz, *Fluid Mechanics*, Course of Theoretical Physics Vol. 6 (Pergamon Press, Oxford, 1987), 2nd ed.