

Dynamical Compartments in Stirred Tank Reactors and Markov State Modeling for Mixing Quantification: A Transfer Operator Approach

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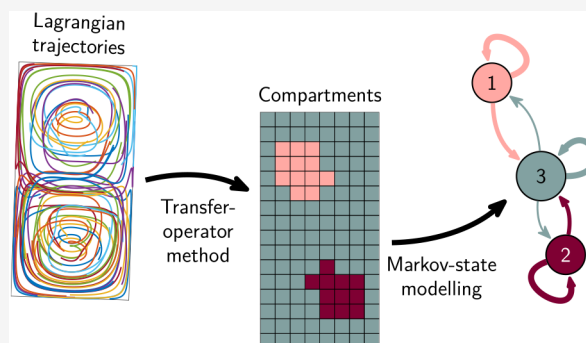


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ABSTRACT: Identifying coherent flow structures in chemical reactors is crucial for understanding the mixing dynamics, which is essential for optimizing reactor performance. We demonstrate the use of a transfer operator method to find coherent flow structures such as almost-invariant sets and coherent sets, which are characterized by minimal mixing with the surrounding fluid, in a lab-scaled stirred tank reactor using both simulated and experimental Lagrangian trajectory data. The proposed method further enables a detailed analysis of the mixing behavior by computing expected residence times and mixing times. Additionally, a Markov state model describes the macroscopic transport dynamics between compartments in the reactor.



1. INTRODUCTION

Understanding and modeling the complex interactions between chemical reactions and fluid dynamics are key in the design and analysis of large-scale chemical reactors. Basic reactor models are global models that are based on reaction kinetics but assume the underlying fluid flow to be either perfectly mixed, ignoring spatial heterogeneities in substrate concentration, or completely segregated, observing each fluid element as a perfectly mixed stirred tank reactor without interaction with the neighboring fluid elements.¹ In reality, a reactor is neither and the truth lies somewhere between these two assumptions² as a result of the interplay between efficiently mixed regions, dead zones, and bypasses. Being aware of these features is highly important for the design of resilient reactors and as such, in the development of SMART reactors in the Collaborative Research Centre 1615 “SMART Reactors for Future Process Engineering”, funded by the German Research Foundation.

Computational Fluid Dynamics (CFD) is increasingly being used to simulate fluid dynamical processes at a high resolution to gain detailed insights into flow fields, including coherent flow patterns and mass transfer. Despite being very useful, CFD simulations are computationally demanding and have their limits when long-term studies of large-scale systems are carried out. The inclusion of reaction kinetics increases the

complexity of the system even more and often leads to numerical instabilities.

Compartment models (CMs) build a bridge between basic reactor models and CFD. The reactor is considered as a network of ideally mixed zones with prescribed measured or precomputed exchange flows between these volumes, see Jourdan et al.³ for a recent review. In this way, CMs form a powerful framework that combines the advantages of local and global approaches: it is computationally feasible but nevertheless takes into account macroscopic heterogeneities in substrate concentrations resulting from limited global mixing. The major issue in the design of CMs is the determination of the compartments and the corresponding exchange flows between the compartments. Typically, CMs are derived from average or steady-state Eulerian flow field analysis, like average or mean velocity fields, average or mean turbulent quantities, or tracer concentrations.^{3,4} Automatic approaches using CFD snapshots have been proposed^{5–7} with recent methods exploiting unsupervised learning.^{8,9} Axial compartments have

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been defined in an automatic procedure in a 580 L stirred tank reactor¹⁰ as well as in a 600 m³ bubble column,¹¹ with the axial fluxes between the compartment volumes estimated by flow following sensors.

As outlined above, a detailed insight into transport processes in the underlying flow field is key for the derivation of CMs. In fluid mechanics, these can be described from the Eulerian or Lagrangian point of view. Eulerian approaches consider instantaneous or time-averaged velocity fields to identify flow structures and compute exchange flows between different reactor volumes in terms of fluxes. The Lagrangian perspective, however, is directly linked to material transport as described by tracer trajectories and thus allows for a more precise, physically meaningful quantification of complex spatiotemporal transport patterns and pathways, which typically do not correspond to those identified by Eulerian methods, especially, when the Eulerian fluxes are merely derived from average fields as in the above cited literature.

To the best of our knowledge, none of the CM frameworks to date define compartments and quantify the exchange flows based on temporally resolved Lagrangian trajectory information, which may be obtained from simulations but also from experimental time-resolved particle tracking in all three spatial dimensions (4D-PTV).^{12,13} A very recent paper also tries to incorporate the Lagrangian aspect of the task to derive the compartments.⁹ In the cited work they compute the compartments in a stirred tank reactor (STR) directly from the concentration distributions of a passive tracer evolved in a frozen flow field obtained by averaging a transient flow simulation. Here, our motivation is to develop an even simpler, but also inherently Lagrangian method to obtain these compartments directly from the objects that are being transported in the transient flow field, namely the Lagrangian trajectories of fluid parcels. The large advantage of our proposed approach in contrast to the former work⁹ is that for the mixing time experiments no additional CFD simulation for the tracer concentration evolution has to be performed but rather, every possible initial position of the tracer feed can be used for mixing time calculations with negligible additional effort as will be shown in the course of this work.

Lagrangian analysis was initially developed and is still in use for the study of transport patterns in geophysical flows.^{14,15} The concepts have been used to, e.g., explain plankton blooms before Madagascar¹⁶ and oil spill dynamics.^{17,18} Only recently these concepts were extended to chemical engineering.^{19,20} Numerous mathematical and computational methods originating from dynamical systems theory have been proposed to study Lagrangian transport and mixing processes^{21–25} building on different, but related notions and definitions of coherent flow structures. We will focus on the concept of almost-invariant and finite-time coherent sets.^{26–29} These are regularly shaped fluid volumes that only weakly mix with their surroundings. Spatially fixed regions in phase space with low escape probability, known as almost-invariant sets, were introduced within a probabilistic approach based on transfer operators.²⁶ This ergodic theoretical concept was extended to the time-variant setting to identify coherent sets, which are mobile.^{27,28,30} The boundaries of such coherent regions can be identified within the geometric approach of Lagrangian coherent structures, where minimal curves or surfaces surround coherent sets.¹⁴

When approximating the transfer operator within a set-oriented numerical framework, a stochastic transition matrix of

a finite-state Markov chain is obtained, which provides the basis for Markov state models and empirical mixing studies. For the identification of the sets of interest, leading eigenvectors of the transition matrix are utilized. Extended methods address the study of almost-invariant and coherent sets that evolve, emerge or disappear^{31,32} and of bifurcations.^{33,34} Moreover, the transfer operator approach was used to model and quantify mixing of two fluids.³⁵ Recent data-based methods make use of unsupervised learning such as spatiotemporal clustering algorithms to extract coherent sets directly from Lagrangian trajectory data.^{36–40}

In Weiland et al.⁴¹ finite-time coherent sets were studied for the first time in the context of chemical process engineering, based on a Lattice Boltzmann simulation of a 2.8 L lab-scale STR. The dominant identified coherent flow structures were derived from Lagrangian tracer trajectories by spectral clustering and were found to be internally mixing. Such coherent sets were tracked and observed in longer time intervals and basic compartment model was derived that took into account the approximate location of the identified compartments and obtained the exchange rates between the compartment volumes from CFD.⁴² Finally, in Steuwe et al.¹³ the coherent set computation from Weiland et al.⁴¹ was repeated on experimentally measured data from 4D-PTV in a corresponding 3 L STR, leading to results that are very similar to the simulated data.

In this contribution, we will push forward the Lagrangian studies of simulated and experimentally measured tracer trajectories in STRs with the perspective of designing a new generation of data-driven compartment models. We will apply for the first time the classical transfer operator-based approach for the identification of almost-invariant and finite-time coherent sets in a process engineering setting. Here, based on a grid discretization of the domain, we obtain the numerical transfer operator as a transition matrix of a finite-state Markov chain, where the transition probabilities are estimated by means of the tracer trajectories. The sets of interest, which form dynamical compartments, are identified from leading eigenvectors of that matrix. This allows for a Lagrangian determination of exchange rates and volume fluxes between those specified dynamical compartments. Additionally, mixing statistics and mixing times can be derived from the stochastic matrix. Finally, mixing time evaluations can be performed repeatedly choosing various possible initial concentration configurations with very moderate computational costs.

The remainder of the paper is organized as follows. In Section 2 we introduce the mathematical and computational background for our Lagrangian framework. After defining different notions of coherent sets that can serve as dynamical compartments (Section 2.1), we review the transfer operator-based approach and demonstrate how the corresponding Markov chain description of the underlying dynamics can be utilized for deriving transport and mixing quantities (Section 2.2). The data sets for our computational studies, comprising both simulated and experimentally measured tracer trajectories from a lab-scale stirred tank reactor with two Rushton turbines and three baffles, are described in Section 3. In Section 4 we present the results of our computational studies regarding the resulting compartments and Markov state models as well as mixing statistics. We conclude with a discussion and an outlook on current and future work in Section 5.

2. THEORETICAL BACKGROUND AND METHODS

In the following, we describe the mathematical and computational background for coherent flow structures in a Lagrangian setting. We will sketch the general framework in the context of nonautonomous dynamical systems. To this end, let $M \subset \mathbb{R}^3$ be the physical space (in our setting the reactor domain), which we assume to be a compact subset of $\mathbb{R}^3$. We consider a time-dependent ordinary differential equation

$$\dot{\mathbf{x}} = \mathbf{u}(\mathbf{x}, t) \quad (1)$$

with state (or position) $\mathbf{x} \in M$, time $t \in \mathbb{R}$, and right-hand side $\mathbf{u}$, which, in our setting, corresponds to a velocity field in physical space and describes the advective motion of passive tracers in an incompressible fluid flow, i.e., volume is preserved. $\mathbf{u}$ is assumed to be sufficiently smooth so that the flow map $\Phi: M \times \mathbb{R} \times \mathbb{R} \rightarrow M$ exists. Given a tracer position $\mathbf{x}$ at time t , evaluating the flow map $\Phi(\mathbf{x}, t; \tau)$ gives its new position after a flow time τ .

Note that in practice, the flow map Φ itself is not explicitly available, so we have to work with N different Lagrangian tracer trajectories $(\mathbf{x}_i(t))_{t \in \mathbb{T}} \in \mathbb{R}^d$, $i = 1, \dots, N$. These may be obtained from the numerical simulation of (1) or from experimental time-resolved particle tracking (4D-PTV). Such trajectories are only given for discrete measurement or evaluation times $t \in \mathbb{T} = \{t_0, t_1, \dots, t_T\}$. We consider uniform time-steps h , i.e., $t_{k+1} = t_k + h$, $k = 0, \dots, T-1$, because this reflects the order of the data sets that are used in this study. Furthermore, for the derivation of the methods, we also assume for now that we do not have any gaps in observation. Taking into account the flow map Φ , we obtain such time-discrete tracer trajectories $(\mathbf{x}_i(t))_{t \in \mathbb{T}}$, $i = 1, \dots, N$ from

$$\mathbf{x}_i(t_{k+1}) = \Phi(\mathbf{x}_i(t_k), t_k; h)$$

So, the given trajectories $(\mathbf{x}_i(t_k))_{t_k \in \mathbb{T}}$, $i = 1, \dots, N$, can be interpreted as the output of iterating the flow map for the N initial conditions $\mathbf{x}_i(t_0)$.

2.1. Almost-Invariant and Finite-Time Coherent Sets

We are interested in the identification of coherent flow structures in the Lagrangian frame of reference. Here, these structures are regular fluid volumes that are characterized by minimal mixing with the surrounding fluid. In the context of nonautonomous dynamical systems, such flow structures correspond to invariant, almost-invariant or finite-time coherent sets.

A set $A \subset M$ is called **Φ -invariant** over $[t, t + \tau]$ if $\Phi(A, t; s) = A$ for all $0 \leq s \leq \tau$. That is, the set (or fluid volume) A remains unchanged under the evolution of Φ , i.e., it does not move as a whole but it might be internally mixing.

Almost-invariant sets obey an approximate invariance principle $\Phi(A, t; s) \approx A$ for all $0 \leq s \leq \tau$.²⁶ To be more precise, given the Lebesgue measure (volume measure) l on M , we call a set $A \subset M$ with $l(A) \neq 0$ **almost-invariant** over the time span $[t, t + \tau]$ if

$$\rho(A) := \frac{l(A \cap \Phi(A, t; \tau))}{l(A)} \approx 1 \quad (2)$$

If $A \subset M$ is almost-invariant over the interval $[t, t + \tau]$, then the proportion of A that gets mapped to A under the action of the flow map is close to 100%. This means that a trajectory that started in A remains in A over the time-span $[t, t + \tau]$ with

high probability. In the Eulerian sense, an almost-invariant set can be viewed as a fixed control volume with minimal in- and out-flow but measured in a Lagrangian manner in terms of the flow map instead of instantaneous fluxes.

Unlike almost-invariant sets that are fixed in phase space, **finite-time coherent sets**²⁸ are allowed to move under the evolution of the time-dependent system. The goal here is to find pairs of sets (or fluid volumes) $(A_t, A_{t+\tau})$, so that tracers starting in A_t at time t will be mapped to $A_{t+\tau}$ with high probability, i.e., $A_{t+\tau} \approx \Phi(A_t, t; \tau)$. This translates into

$$\rho(A_t, A_{t+\tau}) = \frac{l(A_t \cap \Phi(A_{t+\tau}, t + \tau; -\tau))}{l(A_t)} \approx 1 \quad (3)$$

where $\Phi(A_{t+\tau}, t + \tau; -\tau)$ is a set that is obtained from mapping the set $A_{t+\tau}$ at time $t + \tau$ back to time t . Hence, equation (3) measures the proportion of the set A_t at time t that is mapped to the set $A_{t+\tau}$ at time $t + \tau$. You can interpret a coherent pair as a fluid volume that is observed as A_t at time t and as $A_{t+\tau}$ at time $t + \tau$, so that a large proportion of the tracers found in A_t have moved to $A_{t+\tau}$ under the action of the flow and only very little tracers have been lost to other fluid volumes. A related interpretation is that if A_t is an initial blob of high concentration of a chemical substance, then $A_{t+\tau}$ exhibits still most of this initial concentration.

Obviously this concept is ill-posed, because any pair $(A_t, A_{t+\tau})$ with $A_{t+\tau} := \Phi(A_t, t; \tau)$ will be perfectly coherent in the sense that by definition all tracers in A_t will be mapped to $A_{t+\tau}$, obtaining a maximal invariance ratio of 1 in (3). To obtain well-posed problems for finding optimal sets (i.e., fluid volumes) that maximize the ratios (2) or (3) corresponding to almost-invariance and finite-time coherence, respectively, some regularity and mass constraints have to be imposed. These require that the sets of interest are “regular” in the sense that they are stable under small perturbations such as diffusion.^{26,28} This will be ensured by our computational framework approach, which includes numerical as well as explicit diffusion (see Section 2.2). This robustness with respect to perturbation means in our case, that scalar quantities such as concentrations would stay contained in the coherent fluid volumes in the sense that there will only negligible advective-diffusive transport to the surrounding fluid. Thus, the almost-invariant and finite-time coherent sets considered here are fluid volumes that are transported and maybe slightly deformed and possibly well-mixed internally but that resist mixing with the other fluid initially outside of this volume.

We will not focus on the analytical constructions^{28,43} in this work, but rather on data-driven, computational and applied aspects of coherent flow structures, especially in process engineering problems. In the following, we revisit the set-oriented numerical framework for approximating almost-invariant and finite-time coherent sets. We adapt this framework in view of the given simulated or measured trajectories.

2.2. Set-Oriented Method Based on Transfer Operators

As we are interested in the transport of sets and densities under the action of the flow map, we now review a corresponding mathematical concept that is tailored to this problem. We consider the transfer or Perron-Frobenius operator $\mathcal{P}_{t,\tau}: L^1(M, l) \rightarrow L^1(M, l)$ associated with the flow map Φ . As our system is assumed to be incompressible, this linear operator is simply defined by

$$\mathcal{P}_{t,\tau} f(\mathbf{x}) = f(\Phi(\mathbf{x}, t + \tau; -\tau)) \quad (4)$$

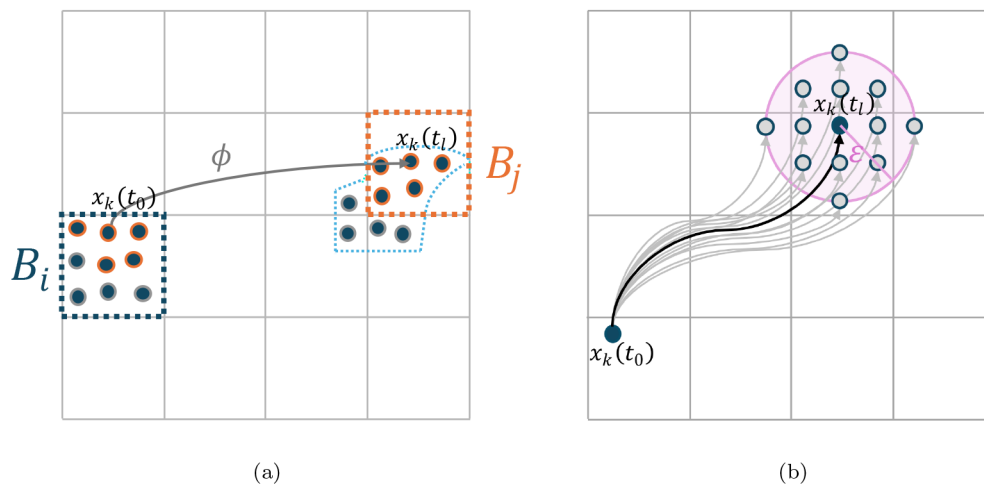


Figure 1. (a): Schematic representation of the transition matrix construction according to equation (7): Each entry P_{ij} is estimated as the fraction of particles from B_i that end in B_j (circled in orange). (b): Introducing diffusive duplicates of each tracer at the final time step within an ϵ -ball on a uniform grid.

The interpretation of (4) is that if f is a density (e.g., representing a scalar field like a concentration field on M) and $f(\mathbf{x})$ the density value at tracer position $\mathbf{x}$ at time t , then $\mathcal{P}_{t,\tau}f(\mathbf{x})$ describes the density value in $\Phi(\mathbf{x}, t; \tau)$ at time $t + \tau$ induced by the flow map. Maximizing ρ in (2) and (3) can be described in the framework of optimizing an inner product involving a compact self-adjoint operator obtained from $\mathcal{P}_{t,\tau}$ in (4).^{28,43} We refer to these works for the functional-analytical background.

We assume for all that follows that our domain M is compact and invariant, i.e., no trajectories will leave M . This is a matching assumption given that the trajectory data studied here originates from simulations and measurements of a closed system. However, the method can be extended to deal with noninvariant subsets of M ,^{28,43,44} which is useful for the future consideration of continuous reactor operations of the STR (CSTR).

2.2.1. Numerical Approximation of Transfer Operators. Let $\{B_1, \dots, B_n\}$ be a partition of M , this is a set of n many small subdomains of the reactor domain so that $\bigcup_{i=1}^n B_i = M$. Here, the partition elements are Cartesian grid cells ("boxes"), but other settings are possible. Applying Ulam's method⁴⁵ a finite-rank approximation of $\mathcal{P}_{t,\tau}: L^1(M, l) \rightarrow L^1(M, l)$ is given via the transition matrix P with entries

$$P_{ij} = \frac{l(B_i \cap \Phi(B_j, t + \tau; -\tau))}{l(B_j)}, i, \dots, n \quad (5)$$

where we drop the t and τ -dependence of P for brevity. In practice, we count how many of the particles released in B_i end in B_j . For this, we consider uniformly distributed sample points $\mathbf{z}_{i,r}$, $r = 1, \dots, R$ chosen in each partition element B_i , $i = 1, \dots, n$. The entries P_{ij} of the transition matrix P in (5) are then estimated via

$$P_{ij} \approx \frac{\#\{r: \Phi(\mathbf{z}_{i,r}, t; \tau) \in B_j\}}{R} \quad (6)$$

where $\#$ denotes cardinality, i.e., the number of elements in a set. P is a sparse, row-stochastic matrix and thus all its eigenvalues are contained in the unit circle. For efficient computation of the transition matrix P we use the GAIO library.⁴⁶

The dynamics induced by P can be interpreted as follows: if $\mathbf{p} \geq 0$ (component-wise) is a density vector (e.g., containing the values of a scalar quantity, such as a concentration, for each box), then $\mathbf{p}' = \mathbf{p}P$ is the push-forward of $\mathbf{p}$ under the discretized action of the flow map $\Phi(\cdot, t; \tau)$ and the vector $\mathbf{p}'$ describes the evolved values after a time span τ . Since P is row-stochastic, the total sum of the entries of $\mathbf{p}$ is preserved under this evolution. In particular, if $\sum_i p_i = 1$, then $\mathbf{p}$ is a

probability vector, and $\mathbf{p}'$ remains a probability vector. Note that the numerical scheme introduces diffusion in the order of magnitude of the box sizes, which is also theoretically needed for robust results.^{28,43}

To construct the numerical transfer operator from given time-discrete tracer trajectories $(\mathbf{x}_i(t))_{i \in \mathbb{T}}$, $i = 1, \dots, N$, $\mathbb{T} = \{t_0, t_1, \dots, t_T\}$, where $t_{j+1} = t_j + h$, one has to adapt (6) as follows. First one has to fix a flow time $\tau = l \cdot h$ as a multiple of h , which is the time step of the trajectory data, and considers the respective interval $[t_0, t_l]$ with $t_0, t_l = t_0 + \tau \in \mathbb{T}$. Let $I_i \subset \{1, \dots, N\}$ be the R_i indices of those initial conditions that can be found in box B_i , i.e., $\mathbf{x}_k(t_0) \in B_i$, $k \in I_i$. P_{ij} then gives the proportion of these initial conditions that will be mapped to box B_j (see Figure 1a for an illustration):

$$P_{ij} \approx \frac{\#\{k \in I_i: \mathbf{x}_k(t_l) \in B_j\}}{R_i} \quad (7)$$

The given data may pose serious constraints. Often the trajectories are nonuniformly distributed and the number of data points per box at the initial and/or the final time step is not sufficient to represent the transition probabilities well. Although the computational framework already incorporates numerical diffusion in the order of magnitude of the box size, we can smooth our construction by introducing additional diffusion. A simple way is to consider a multivalued mapping approach: We introduce N_{diff} diffusive duplicates of each tracer $\mathbf{x}_k(t_l)$ at the final time step t_l . For this we generate a grid in an ϵ -ball (a ball with radius ϵ) around each $\mathbf{x}_k(t_l)$, depicted in Figure 1b. The N_{diff} grid points are then positions of pseudo tracers at time t_l with the same initial position $\mathbf{x}_k(t_0)$ as the original tracer. Let R_i be the number of tracers starting in box B_i and $\hat{R}_{ij}^\epsilon$ the number of diffused pseudo tracers starting in box B_i and ending in box B_j . The pseudo tracers around each real tracer particle start in the same location as the real particle but end up at slightly moved locations, mimicking a diffusive process. Then we get the following diffused transition matrix

$$P_{ij}^{\text{diff}} \approx \frac{\hat{R}_{ij}^\epsilon}{N_{\text{diff}} \cdot R_i} \quad (8)$$

To study the transport over a series of adjacent time intervals $T_1 = [t_0, t_1]$, $T_2 = [t_1, t_2]$, ..., $T_f = [t_{(f-1)h}, t_{fh}]$ of length $\tau = l \times h$, where $t_{k+l} \in \mathbb{T}$ for $k = 0, \dots, f$, we can construct a family of transition matrices $P_{t_0, \tau}, P_{t_1, \tau}, \dots$ over the respective time intervals. For a time-dependent velocity field $\mathbf{u}$, these matrices are different in general. When the system is autonomous, i.e., $\mathbf{u}$ is time independent, $P_{t_k, \tau}$ only depends on the length τ of the intervals and not on the initial times. Therefore, any two transition matrices $P_{t_i, \tau}, P_{t_j, \tau}$ computed w.r.t. to initial times

$t \neq s$ are then identical. Further, when u is a time-dependent periodic velocity field with period τ , that is, $u(x, t) = u(x, t + \tau)$, any two transition matrices $P_{t,\tau}$ computed w.r.t. to initial times t and $s = t + k \times \tau$, $k \in \mathbb{Z}$ are identical. For autonomous and periodic systems this can be exploited accordingly, by cutting the given trajectories into segments of length τ and interpreting all of these short segments as virtual trajectories on a single time interval, that is, on $T_1 = [t_0, t_f]$. This such enriched trajectory data set on T_1 is the basis for constructing a single transition matrix, which we will term “combined transition matrix for one stirrer rotation”. This concept can also be used for an approximately periodic flow.

2.2.2. Extraction of Almost-Invariant and Finite-Time Coherent Sets. The general aim is to find an optimal partition of M into subsets (i.e., compartments) $A_1, \dots, A_{K+1}$, that mix little with the rest. This processing step can also be understood as a means of finding the best number of compartments and their shape in order to represent the dynamic system of transport in a CM model. The number and shape of the ideal compartmentalization is current subject of study.⁹ In our computational framework each A_j , $j = 1, \dots, K + 1$, is made up of boxes, i.e., $A_j = \bigcup_{i \in I_{A_j}} B_i$, where I_{A_j} denotes the set

of indices $I_{A_j} \subset \{1, \dots, n\}$ of the grid cells that form A_j . We obtain such a partition based on the numerical transfer operator P .

In a nutshell, the transition matrix P is a stochastic matrix, with eigenvalues contained in the unit circle. In practice, we will form a transition matrix R of a reversible Markov chain from P ,⁴³ which exhibits a real-valued spectrum. The leading eigenvalue λ_1 is always 1 and the corresponding eigenvector v_1 is a vector of ones, while all further eigenvectors to eigenvalues are orthogonal to v_1 and thus contain signed entries. One would find a nearly optimal partition of M into two almost-invariant sets from the sign structure of the second eigenvector v_2 to the eigenvalue $\lambda_2 < 1$ (but close to 1) by defining $A_1 = \bigcup_{\{j: v_{2,j} \geq 0\}} B_j$ and $A_2 = \bigcup_{\{j: v_{2,j} < 0\}} B_j$.²⁶ In general, almost-invariant

sets are extracted from several eigenvectors $v_2, \dots, v_K$ of this transition matrix R obtained from P to eigenvalues $1 > \lambda_2 \geq \dots \geq \lambda_K$, where K is chosen such that $\lambda_K - \lambda_{K+1}$ is large (i.e., there is a spectral gap). Similarly, for finite-time coherent sets singular vectors of P or a related matrix are considered. We state the exact algorithms in Appendix A. A strong improvement in the capturing of the mass transfer dynamics by adding further compartments can only be expected when the eigengap between successive eigenvalues is high.

To extract almost-invariant and coherent sets from the corresponding vectors $v_2, \dots, v_K$, we postprocess these vectors to disentangle the sets using the SEBA (Sparse EigenBasis Approximation) algorithm:⁴⁷ The basis $V := (v_1, \dots, v_K)$ of the eigenspace $\mathcal{V} \in \mathbb{R}^n$ is transformed into a sparse basis $S := (s_1, \dots, s_K)$ of the vector subspace $\mathcal{S}$, such that $\mathcal{V} \approx \mathcal{S}$. The entry s_{ij} of the matrix S gives the likelihood of box B_i , $i = 1, \dots, n$ to belong to the box-valued sets A_j , $j = 1, \dots, K$ as defined above. By choosing a threshold, each box B_i can be assigned to a specific compartment A_j or to the incoherent background, which will form the additional compartment A_{K+1} , respectively.⁴⁷ The vector $s_{\max} \in \mathbb{R}^n$, which describes an overall cluster likelihood for each particle with entries $s_i = \max_j(s_{ij})$, $i = 1, \dots, N$, $j = 1, \dots, K$ can also be considered.

2.2.3. Markov Chain Statistics. Using a transition matrix P , we can evolve any (box-discretized) initial distribution in time. In particular, given a family of transition matrices over adjacent time intervals, $T_1, T_2, \dots, T_f$ we can push forward an initial density vector p via $pP_{t_0,t_1}P_{t_1,t_2}\dots P_{t_{f-1},t_f}$. The transfer operator over a long time span can thus be approximated by multiplication of transition matrices computed over shorter time intervals. In practice, only consecutive matrix-vector multiplications with sparse transition matrices have to be carried out, which can be computed very efficiently. Hence, fluid mixing can be described and quantified (e.g., by the variance or other mixing measures) using a numerical transfer operator P .³⁵

In the following, we assume that we have a time-homogeneous Markov chain with transition matrix P , corresponding to a time-periodic or autonomous velocity field. We consider a set A consisting

of boxes B_i , $i \in I_A$, where $I_A \subset \{1, \dots, n\}$ is the set of indices such that $\bigcup_{i \in I_A} B_i = A$. We can compute the **expected residence times** for A , the expected time to stay in A before leaving and reaching any other part of the domain $M \setminus A$. Let P_A be the transition matrix P restricted to the states in A . We assume that A is transient, i.e., eventually all mass initialized in A will leave A under the repeated action of P_A . The expected residence time to stay in A when starting in box $B_i \subset A$ is then given as

$$r_i = \sum_{j \in I_A} (I - P_A)^{-1}_{ij}$$

In practice, this can be computed by solving

$$(I - P_A)r = \mathbf{1} \quad (9)$$

where the vector r contains the entries r_i , and $\mathbf{1}$ denotes the all-ones vector.

For a given initial density vector v (for example an initial blob of high concentration), the **mixing time** describes how long it takes for the system to approach equilibrium within a defined tolerance. The vector of equilibrium values is obtained via the invariant distribution of the Markov chain, which is given by the left eigenvector π of P to the leading eigenvalue $\lambda = 1$, normalized to match the total concentration. Although our flow is volume-preserving, the invariant distribution vector is not uniform because some boxes only partially intersect the interior of the reactor. These smaller effective volumes lead to the corresponding eigenvector entries in π being smaller than those with respect to boxes entirely within the reactor. Our approach thus respects the numerically (slightly) nonuniform invariant distribution when computing the mixing times below.

We evolve the concentration vector v until 95% of all boxes have a concentration value within $\pm 5\%$ of their corresponding equilibrium value and define the number of time steps required as the mixing time. In the following, both the expected residence time and mixing time are given in number of discrete time steps of length τ .

Further, we can set up a **Markov state compartment** model by considering the transitions between almost-invariant sets and the surrounding region. Suppose you are given the $K + 1$ disjoint sets of interest $A_1, \dots, A_{K+1}$ that are composed of boxes and $\bigcup_{k=1}^{K+1} A_k = M$. Let $I_{A_1}, \dots, I_{A_{K+1}}$ denote the sets of indices such that $\bigcup_{i \in I_{A_k}} B_i = A_k$, $k = 1, \dots, K + 1$. Note that we think of K almost-invariant sets plus the remainder of the domain, which will form the set A_{K+1} . The transition matrix $\bar{P} \in \mathbb{R}^{(K+1) \times (K+1)}$ that describes the coarse grained dynamics between these $K + 1$ compartments is obtained as follows: Let $e_{A_k} \in \mathbb{R}^n$ be an indicator vector of the set A_k , i.e., $(e_{A_k})_i = 1$ for $i \in I_{A_k}$ and $= 0$ otherwise. The entries of $\bar{P}$ are computed via

$$\bar{P}_{ij} = \frac{e_{A_i}^T P e_{A_j}}{\#I_{A_i}}, \quad i, j = 1, \dots, K + 1 \quad (10)$$

In case of finite-time coherent sets as compartments, one considers $K + 1$ different pairs of sets $(A_k, A_{k,\tau})$, $k = 1, \dots, K + 1$, with indicator vectors $e_{A_k,t}$ and $e_{A_{k,\tau},t+\tau}$, respectively, where again the $(K + 1)$ -th pair is identified with the incoherent background. The entries of the coarse-grained transition matrix $\bar{P}_{t,\tau} \in \mathbb{R}^{(K+1) \times (K+1)}$ are then computed from $P_{t,\tau}$ in analogy to (10) as

$$\bar{P}_{ij,t,\tau} = \frac{e_{A_i,t}^T P_{t,\tau} e_{A_j,t+\tau}}{\#I_{A_i,t}}, \quad i, j = 1, \dots, K + 1 \quad (11)$$

This equation gives the conditional probability that a tracer initialized in compartment $A_{i,t}$ at time t can be found in compartment $A_{j,t+\tau}$ at time $t + \tau$. The probability $\bar{P}_{i,j,t,\tau}$ describes the transition from $A_{i,t}$ to $A_{j,t+\tau}$, i.e., the probability to stay in the (time-dependent) coherent compartment, which should be large by construction of coherent sets.

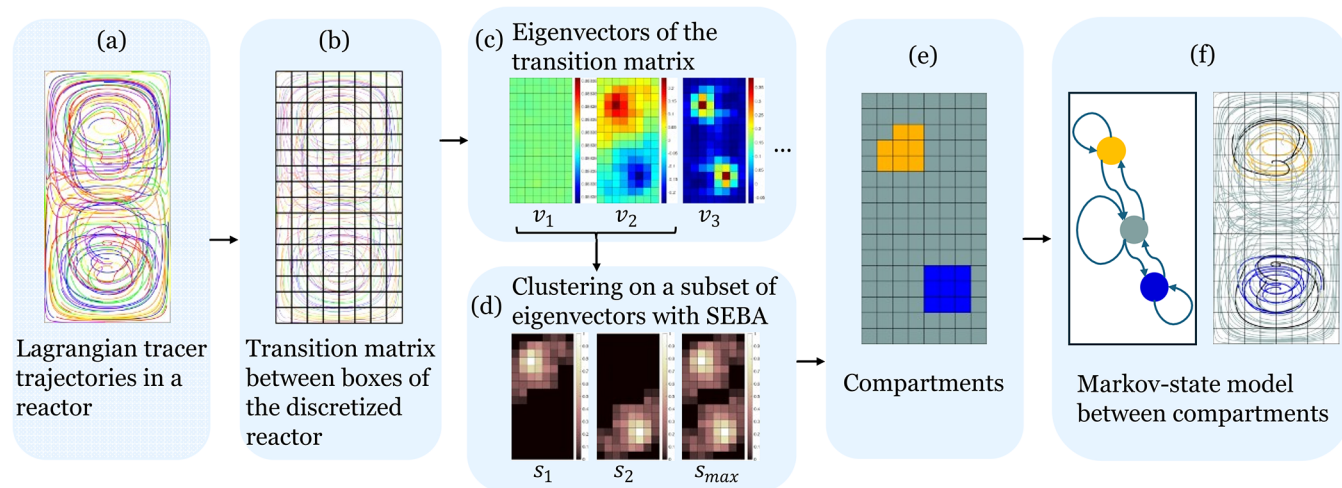


Figure 2. Schematic workflow of the proposed data-driven computational framework. From given tracers in a reactor (a) we compute a transition matrix on the discretized domain (b). The eigenvectors of the transition matrix indicate almost-invariant sets in physical space (c). Using a clustering technique on a subset of leading eigenvectors (d), we can extract compartments (e). Using these compartments, we can build a coarse-grained Markov state model (f). Trajectories that start in a compartment end with high probability in this compartment.

These Markov state models provide simplified representations of the macroscopic dynamics of the system. Typically, there is little transport between different compartments, which is how these sets are characterized from (2) and (3), respectively, while there can be a high level of mixing within individual compartments.

Using the volumes of the compartments, one can also compute the **transition volumes** or volume flow rates Q from $\bar{P}$ (similarly for $\bar{P}_{t,r}$). The entry $\bar{P}_{ij}$ is the conditional transition probability from compartment A_i to A_j . The volume flow rate from compartment A_i to A_j is then given as

$$q_{ij} = \frac{\bar{P}_{ij} \cdot I(A_i)}{\tau}$$

where the volume $I(A_i) = \sum_{j \in I_{A_i}} I(B_j)$ can be straightforwardly computed from the side lengths of the boxes B_j . If the geometric volumes of the boxes B_j are only approximate (for example, if the boxes are cubic and extend partially outside the actual reactor), one can correct the compartment volumes using the leading left eigenvector π of P , normalized to match the total volume. One estimates the volume of compartment A_i then as $\tilde{I}(A_i) = \sum_{j \in I_{A_i}} \pi_j I(B_j)$.

Summary of the Numerical Framework. In this section, we have introduced a computational data-driven approach based on transfer operators to study reduced models for transport and mixing processes in chemical reactors based on experimentally measured or numerically simulated Lagrangian tracer trajectories. The schematic workflow is summarized and visualized in Figure 2.

3. STIRRED TANK REACTOR DATA

The trajectory data of the STR originate from both simulations and experiments conducted in a lab-scaled vessel (see Figure 3) filled with water and stirred by two Rushton turbines at a stirrer frequency of 252 rpm. The STR is further equipped with three baffles and a korboggen head bottom.

3.1. Simulated Data

The data results from Lattice Boltzmann (LB) simulations conducted with the program M-Star CFD version 3.7.123. A total of $g = 300$ lattice sites are chosen across the vessel's diameter, resulting in a resolution of $\Delta x = 4.3 \times 10^{-4}$ m. In previous publications,⁴¹ this resolution delivered a grid independent solution, meaning that the results did not change significantly by increasing the resolution beyond that point. To

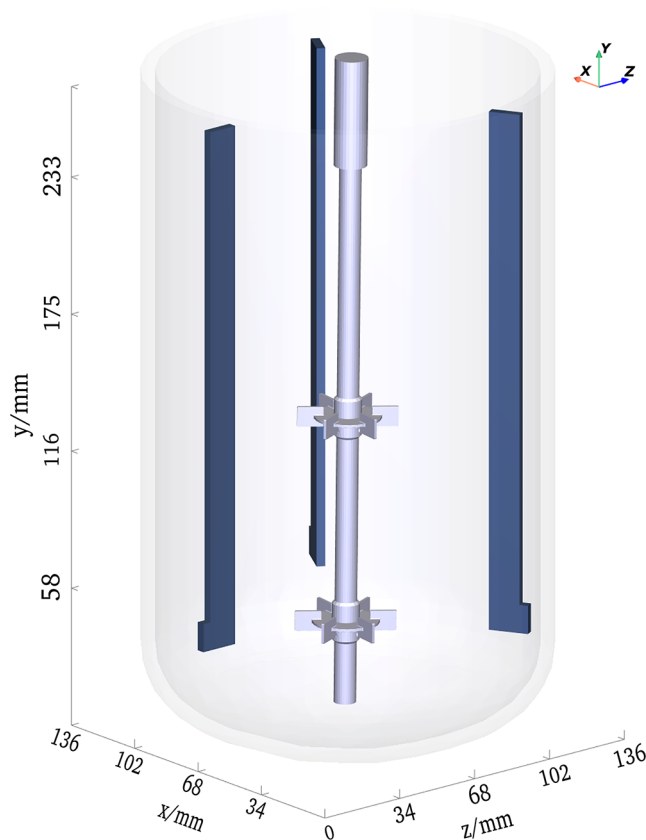


Figure 3. Schematic representation of the stirred tank reactor.⁴¹ Dimensions are length (x): 127.94 mm, width (z): 127.94 mm, height (y): 223.64 mm.

account for turbulent modeling, the simulations are carried out as Large Eddy Simulation (LES). The turbulent viscosity is modeled with the Smagorinsky-Lilly subgrid-scale model⁴⁸ with a Smagorinsky constant of $C_S = 0.1$. The simulation is run for a time of 60 s to achieve a quasi-stationary flow. After this time, the volumetric data concerning the time-dependent velocity field is stored at a frequency of $f_{\text{save}} = 500$ Hz over a period of approximately $t_{\text{save}} = 2.5$ s, covering slightly more

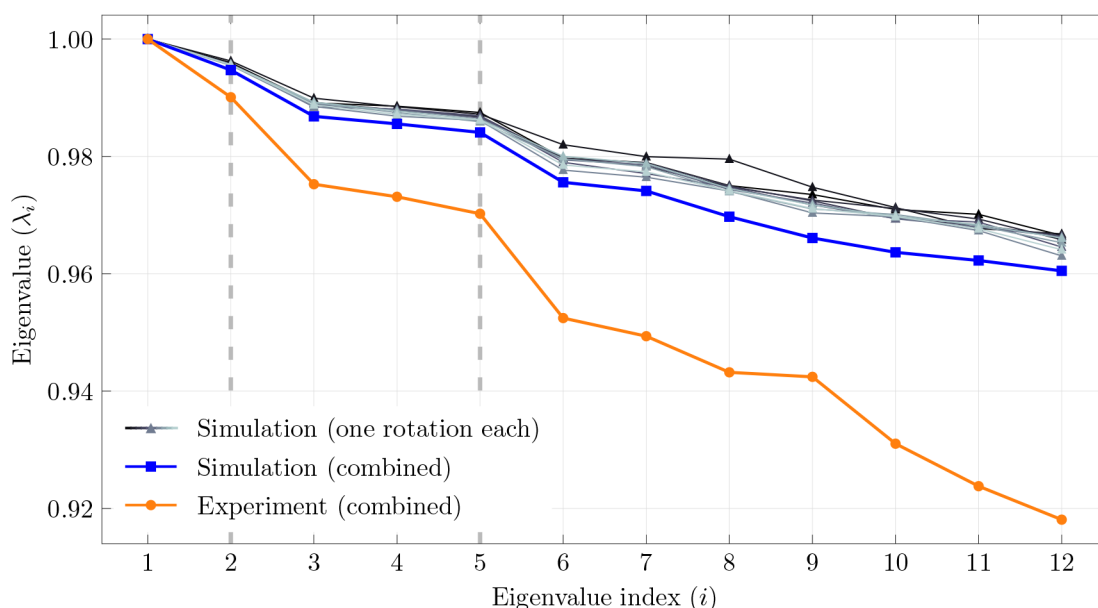


Figure 4. Leading 12 eigenvalues for ten transition matrices (symmetrized, see Appendix A.1) for the time intervals $[0,1]$, $[1,2]$, ..., $[9,10]$ (time expressed in number of rotations) for the simulated data (from darkest shade $\blacktriangle$ for the first rotation to the lightest shade $\triangle$ (blue-green) for the tenth rotation). Eigenvalues of the corresponding combined transition matrices for one stirrer rotation for the simulated ($\square$) (blue) and the experimental ($\circ$) (orange) data.

than ten full stirrer rotations. This data is then used to advect a total of roughly 342,000 particles by the flow to retrieve individual trajectories. A more detailed description of this process is given in previous work.^{41,42}

For validation of the mixing characteristics, additional simulations are carried out to determine the mixing time in the observed STR. For this, at four selected positions a tracer is added after an initialization time of $t_{\text{init}} = 30$ s. The initial tracer positions correspond to those shown in Figure 10. The transport of this tracer is described by the advection-diffusion equation. Over a time of $t_{\text{advect}} = 60$ s the concentration at each lattice site is compared with the mean concentration of the respective tracer in the whole vessel, declaring a site mixed if the local concentration is within a $\pm 5\%$ deviation from the mean concentration. Since Rautenbach et al. have shown that such simulations are not exactly reproducible,⁴⁹ they are executed ten times. The mixing time results are shown in (Figures S8 and S9 Supporting Information).

3.2. Experimental Data Set

The experimental data set originates from a 4D Particle Tracking Velocimetry (4D-PTV) measurement that was carried out and published by Steuwe et al.¹³ 4D-PTV is a novel measurement technique in which particles are tracked in a flow using multiple cameras with different lines of sight, in order to obtain time-resolved reconstructions of the particle tracks in three-dimensional space. From these trajectories the Lagrangian velocity can be derived by differentiation.⁵⁰ In the study by Steuwe et al.,¹³ the experiment was conducted at 22 °C in a handmade glass vessel filled with 3 L of bidistilled water and seeded with approximately 40,000 fluorescent, monodisperse particles measuring 50 μm in diameter. Due to their small size compared to the underlying flow structures and their neutral buoyancy, the particles are considered to behave as passive tracers in the flow.¹³ The experimental setup includes three baffles and two Rushton turbines, operating at 252 rpm. This matches the setup for the simulation data set by Weiland et al.⁴¹ introduced in the previous paragraph. The resulting data set consists of $37,879 \pm 13$ particles per time step tracked over 2,818 time steps at a rate of 500 Hz. This corresponds to a total investigation time of 5.64 s. Since the

experiment was carried out using five high-speed cameras positioned around the entire STR, the data set covers the full reactor volume. Due to the 4D reconstruction algorithm, the tracks are occasionally lost and found again (less than 100 particles are lost and found each time step), resulting in a total of 216,000 individual particle tracks. The loss of particle trajectories must be taken into account when using the analysis method presented here.

4. COMPUTATIONAL RESULTS

In this section we carry out computational analyses in a STR using the simulated and experimental trajectory data as introduced in Section 3, applying the set-oriented framework discussed in Section 2.2. We demonstrate the extraction of compartments (i.e., almost-invariant and coherent sets) using the transfer operator approach within a set-oriented numerical framework. Moreover, we derive Markov state models to describe the macroscopic dynamics and study scalar mixing.

4.1. Compartments and Markov State Model

First, we apply our set-oriented method to ten consecutive stirrer rotations of the simulated data, i.e., $\tau \approx 0.2381$ s (one rotation). We consider the respective parts of the simulated trajectories on the ten different time intervals $[0,1]$, $[1,2]$, ..., $[9,10]$ (time expressed in number of rotations). To set up the transitions matrices, the domain is partitioned into small boxes with side lengths $l_{Bx} = l_{Bz} = 4$ mm and $l_{By} = 3.8$ mm. To compute the transition matrices, we add diffusion (radius $\epsilon = 4$ mm, a regular grid containing $N_{\text{diff}} = 33$ test points) as described in equation 98) in Section 2.2. In the Supporting Information we provide detailed parameter studies that demonstrate the robustness of our results with respect to changes of these computational parameters (see Table S1 and Figures S1–S6). The number of boxes ranges between 47,993 and 49,029 for the different time intervals, since we can only use boxes with available trajectory data.

We use Algorithm A.1 in the appendix and compute the leading eigenvalues of the symmetrized transition matrices (Figure 4, gray curves). For all matrices, there is a clear

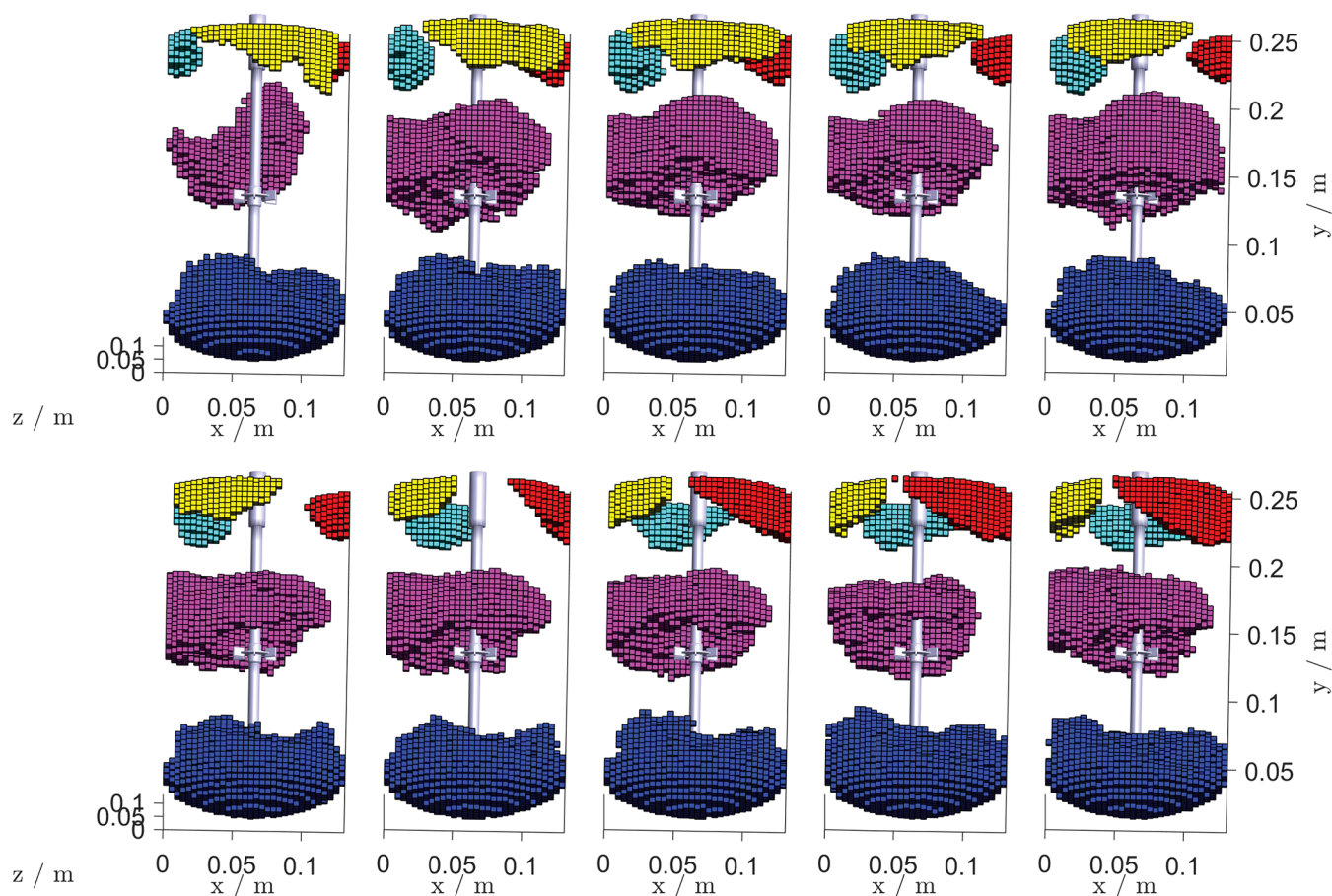


Figure 5. Extracted five almost-invariant sets on time intervals $[0,1]$, $[1,2]$, ..., $[9,10]$ with time expressed in number of rotations.

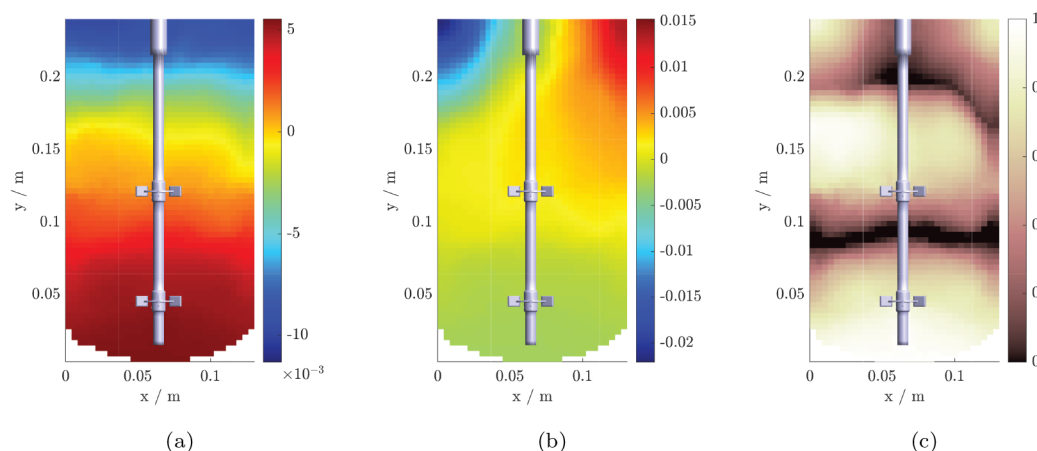


Figure 6. Eigenvectors to the second (a) and fourth (b) eigenvalue of the combined transition matrix for one stirrer rotation (symmetrized matrix R , see Appendix A.1) and $s_{\max}$ (c) serving as a cluster indicator; all shown in the middle plane $z = 0.0635$ m.

eigengap after the fifth eigenvalue indicating the existence of $K = 5$ prominent almost-invariant sets. For some of the matrices there are further spectral gaps, e.g., after the eighth or 11th eigenvalues, but not as clearly. Therefore, further partitioning of the reactor into more compartments is not necessary to grasp the large-scale chaotic transport for the studied STR with two Rushton stirrers. In comparison to other work on STRs^{9,51} the determined number of necessary compartments is very low, however, here the shape of the compartments was determined from the Lagrangian information and not from a frozen flow

field that does not represent the dynamical system for transient flows.

To extract the almost-invariant sets, we consistently use a cut value of 0.7 on the SEBA vectors (postprocessed eigenvectors, see Section 2.2.2, and we will refer to the general scheme again below, see Figure 6).

Figure 5 shows the extracted five almost-invariant sets for ten different stirrer rotations. In each of the ten cases we obtain three smaller regions located at the top, one in the center, and one at the bottom of the reactor, which appear to be very similar in shape and position for the ten different stirrer

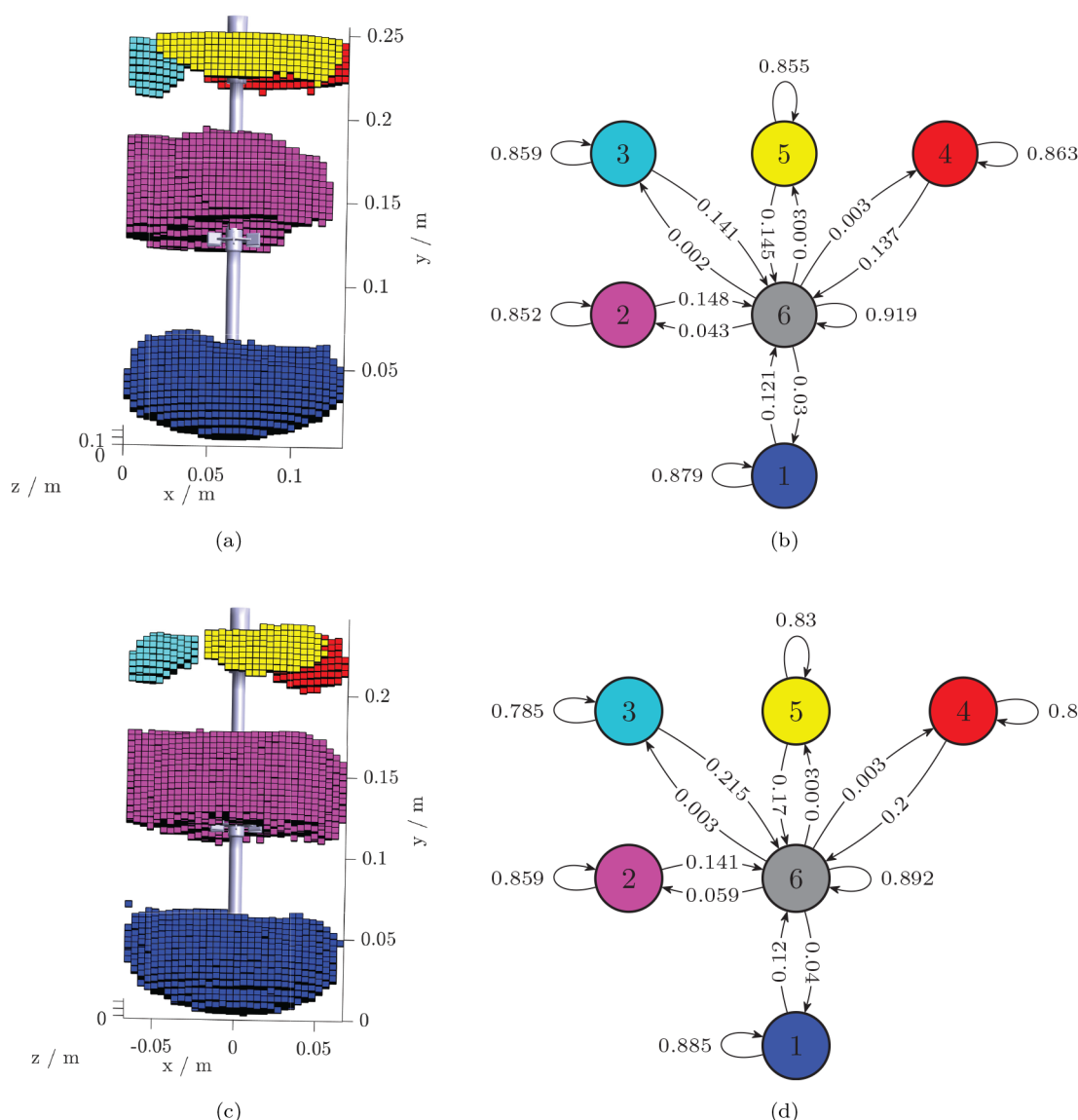


Figure 7. Five extracted almost-invariant sets for the simulated data (a) and experimental data (c) computed from the combined transition matrix for one stirrer rotation. Markov state compartment model using the five almost-invariant sets and the surrounding region as compartments for the simulated data (b) and experimental data (d). The transition diagrams show the transition probabilities between the compartments over one rotation of the stirrer.

rotations. In particular, the bottom set seems to be very robust, whereas one observes a slight rotation of the upper three sets from one time interval to the next.

Assuming the dynamics to be nearly periodic with a period equal to one stirrer rotation, we treat the system as time-periodic, following the approach outlined in Section 2.2.1. In other words, we now combine the information on ten stirrer rotations obtained from the simulated trajectories into a single transition matrix for one stirrer rotation, where 49,983 boxes are used. The actual number of trajectories in each box at initial times ranges from 1 to 133 with median 77. Again, we compute the leading eigenvalues (Figure 4) and corresponding eigenvectors. In Figure 6a we show the eigenvector corresponding to the second largest eigenvalue, that is, we plot the boxes colored by their corresponding eigenvector entry (restricted to the middle $z = 0.0635$ m). The sign structure of this eigenvector appears to divide the reactor into an upper and lower part and this information could be used to

define two almost-invariant sets. However, the subsequent eigenvectors contain information about smaller almost-invariant sets, see Figure 6b, where at the top of the reactor two regions are highlighted by large-magnitude positive and negative entries (shown in red and blue, respectively). Now, to extract almost-invariant sets based on the eigenvectors of the five leading eigenvalues, we use SEBA as described in Section 2.2.2: We obtain five sparse approximate eigenvectors from the original ones, with each one highlighting one single almost-invariant set (corresponding to vector entries close to 1, see also the workflow illustrated in Figure 2d). We plot the entry-wise maximum $s_{\max}$ of all these output vectors (which can be interpreted as an overall cluster indicator) in Figure 6c, by that visualizing the location of the five almost-invariant sets (three at the top, with one out of plane, and one at the center and the bottom, respectively).

We obtain the $K = 5$ almost-invariant sets for the numerical data shown in Figure 7a. The bottom compartment consists of

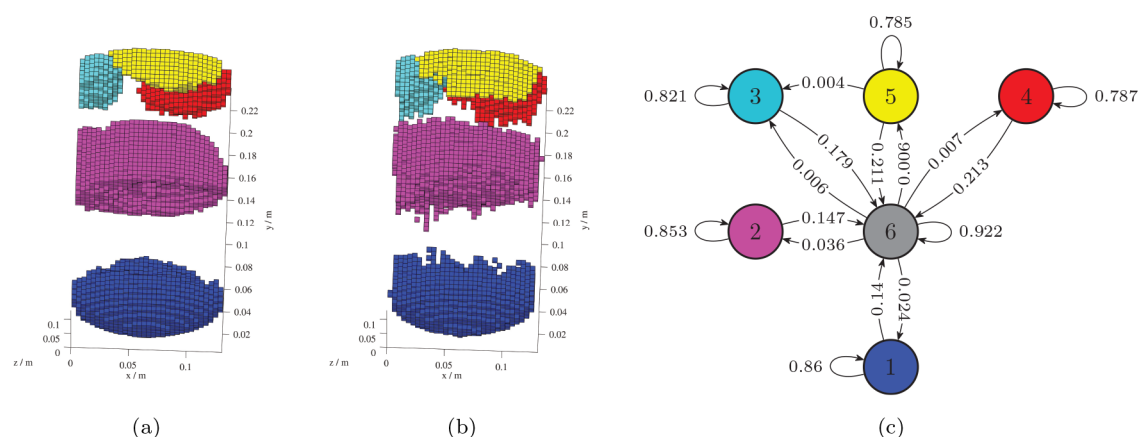


Figure 8. Extracted coherent sets from the combined transition information for one stirrer rotation at initial time (a) and at final time (b), for simulated data. (c): Corresponding transition state diagram for one rotation with the coherent pairs and the background forming the compartments.

8,271 boxes, the center of 8,859 boxes, and the three in the top of 431 (cyan), 793 (yellow) and 637 (red) boxes.

To obtain a simplified representation of the macroscopic dynamics in the tank reactor, we use these five sets (1–5) – along with the surrounding region (6) – as compartments in a Markov state model. Since we defined these compartments using the transition probabilities between the boxes, we can also directly obtain the corresponding transition state probabilities between the six compartments using eq 10. The transition state diagram for the numerical data is shown in Figure 7b. For example, a tracer initialized in the lower compartment 1 (blue) will, with probability 0.879, still be in that compartment after one rotation of the stirrer, while with probability 0.121 it will be found in the incoherent background compartment 6. Expanding this, a tracer starting in compartment 1 might be found in compartment 2 (magenta) with probability $0.121 \times 0.043 \approx 0.005$ after two rotations, following the arrows in the diagram. In this way, we have a reduced stochastic model at hand that describes the macroscopic transport between the six compartments. The transition probabilities in the diagram are low between the different compartments, which means that there is little transport between them. At the same time, the probabilities for the fluid or tracer to stay within its current compartments is very high. This is another demonstration that our identified compartments are almost-invariant.

For the experimental data, not all tracked particles are available at every time frame. To apply our method, we require particles that are available at both the initial and final times of the selected time interval. Here we can use between 29,856 and 32,831 trajectories for each rotation. We again form the combined transition matrix for one stirrer rotation (using all 23 rotations). The reactor domain is partitioned into 44,840 boxes with side lengths $(l_{Bx}, l_{By}, l_{Bz}) = (4.2, 3.6, 4.2)$ mm. The actual number of trajectories in each box at initial times ranges between 1 and 42 with median 18. We add diffusion using an ϵ -ball with radius $\epsilon = 8$ mm, discretized by $N_{\text{diff}} = 360$ grid points. When choosing a smaller diffusion radius (e.g., $\epsilon = 4$ mm as above for the simulated data), the transition matrix has multiple eigenvalues 1, indicating several isolated boxes. Therefore, we use a higher diffusion radius to obtain an eigenvalue $\lambda_2 < 1$.

When comparing the experimental and simulated data, we observe similar spectral gaps after the second and fifth

eigenvalues (Figure 4) as well as another, less pronounced eigengap after λ_9 . For the experimental data, we extract $K = 5$ very similar almost-invariant sets that are only slightly shifted, with minor differences in sizes (Figure 7c). Here, the bottom compartment consists of 8,024 boxes, the center of 10,133 boxes, and the three in the top contain 470 (cyan), 401 (yellow) and 540 (red) boxes, respectively. This shows a consistent compartmentalization of the dynamics across numerical and experimental data. The corresponding transition state diagrams also agree very well (Figure 7d). We note that due to having to select a threshold on the SEBA vectors to extract the sets of interest, there might be isolated boxes that are part of compartments, as in Figure 7c for the bottom compartment. These could be discarded in a postprocessing step. The dependence of the resulting compartments on the cut value is very robust, see Figure S6 in the Supporting Information.

In theory, we can also identify compartments that move and change over time. Figure 8a shows extracted coherent sets for the simulated data using the previously considered combined transition matrix for one stirrer rotation, constructed from the simulated trajectories. For the computation of the coherent sets we used Algorithm A.2 in the appendix, which utilizes singular vectors of the transition matrix. A transition state model is derived using equation (11), see Figure 8b. Note that the compartment pairs in Figure 8a and b differ slightly in shape, with the sets at final time (Figure 8b) a bit larger and more irregular, with some numerical artifacts due to having to select a threshold for extracting the sets, than those at initial time (Figure 8a). However, the positions have hardly changed. Moreover, when comparing the transition state diagram (Figure 8c) with that for the Markov state model based on almost-invariant sets (Figure 7b), it appears that the time-dependent compartments in Figure 8c are even slightly less coherent (compare the transition probabilities within and between compartments). This confirms that our previous assumption of an approximately periodic flow with spatially fixed compartments that can be obtained from almost-invariant sets was reasonable.

From the transition probabilities shown in Figure 7b,d and Figure 8b also the physical volume flow rates or material fluxes can be derived. Therefore, the results presented in this section enable us to derive a reactor network for compartment modeling (CM) directly from Lagrangian data. This ensures

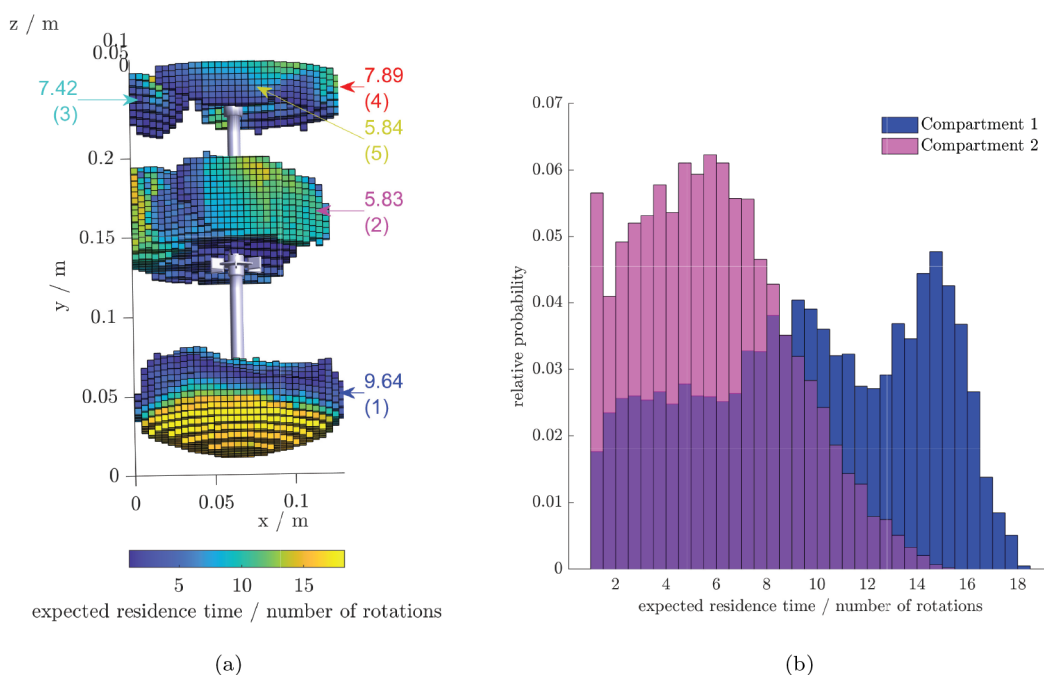


Figure 9. Expected residence times computed from the combined transition information for one stirrer rotation. (a): Color of a box shows the expected residence time for a tracer starting in that box to stay in its initial compartment. The mean expected residence times for compartments (1)–(5) are labeled (in units of stirrer rotations). (b): Histograms of the expected residence times for the middle (magenta) and bottom (blue) compartment.

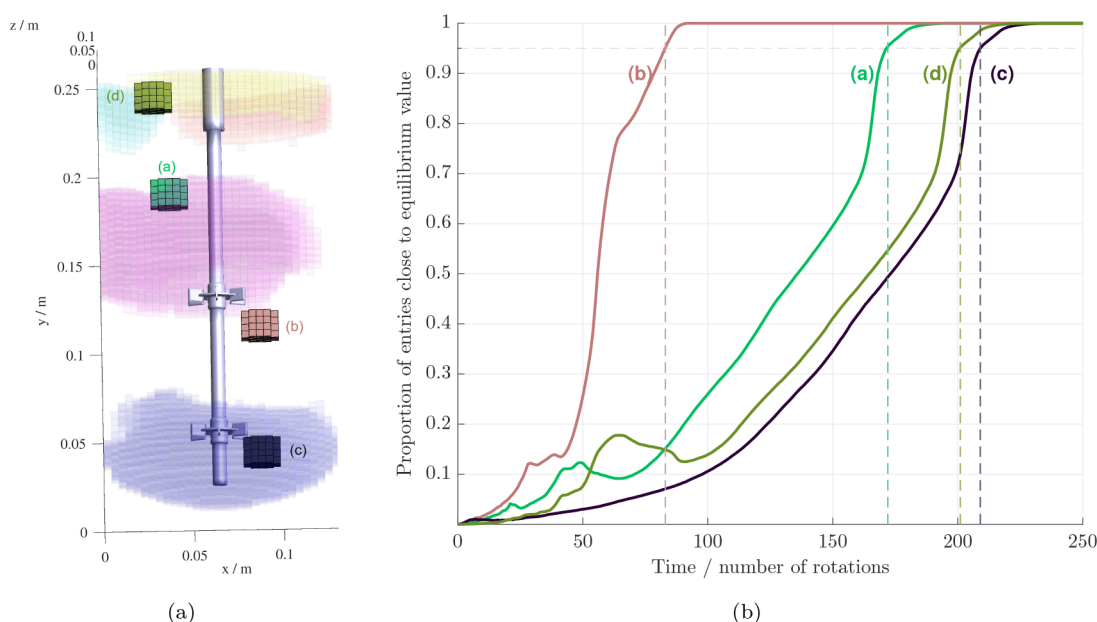


Figure 10. (a): Green (spot a), dusty pink (spot b), dark gray (spot c) and olive green boxes (spot d) show the initialized positive values for the four density vectors. (b): Proportion of vector entries close to the equilibrium value over time for the evolved density vectors. Starting from spot a (green line), spot b (dusty pink line), spot c (dark gray line), spot d (olive green line). The dashed lines mark the mixing times.

that the correct dynamical system for the transported quantities forms the basis for the compartments. We stress that the dynamical system of the mean flow, which is typically used for setting up macroscopic models, might be a very different one.¹⁴ Whether a compartment should then be modeled in a CM by an ideal plug flow reactor or an ideal STR could further be estimated by the internal mixing of the corresponding compartment, which can also be evaluated using our Lagrangian methods. In the future, reactors can be

analyzed based on the Lagrangian method presented here, to provide a CM digital twin. This digital twin could then be used for the evaluation of different reaction pathways without the need of further costly experiments or tedious CFD simulations.

4.2. Mixing Statistics

The computed transition matrix P can further be utilized to compute various mixing statistics. Here, we use only the combined transition matrix for one stirrer rotation of the simulated data to demonstrate these methods.

For each almost-invariant compartment we compute the expected residence times using equation (9) (Figure 9a). This quantity represents the average number of rotations required for a particle in a given box to leave the corresponding compartment. The mean expected residence time for each compartment is highest for the bottom compartment with 9.64 rotations, followed by two of the top sets with 7.89 and 7.42 rotations, respectively. The remaining top and the middle compartment both have lower expected residence times with around 5.8 rotations. Notable, a larger proportion of boxes in the bottom compartment have higher expected residence times than those in the other compartments (see for instance the comparison with the middle compartment in Figure 9b).

We evolve four different initial distributions of a scalar quantity such as a educt concentration. Here, four cylindrical spots are initialized with a uniform density (filled) and zero elsewhere (see Figure 10a). Three of these are deliberately chosen: one is positioned slightly above the middle compartment (spot a), one between the middle and bottom compartments (spot b), and one in the bottom compartment (spot c). The fourth spot, located at the very top (spot d), emulates experimental settings reported in Fitschen et al.^{52,53} with the same stirrer configurations (2x Rushton) and overall geometry. Let $\mathbf{p}_{(x),0} \in \mathbb{R}^n$ (where $n = 49,983$ is the number of boxes and $x \in \{a,b,c,d\}$) be such an initial density vector. This is evolved over time (given in number of stirrer rotations) efficiently by

$$\mathbf{p}_{(x),k+1} = \mathbf{p}_{(x),k} \mathbf{P}, \quad k = 0, 1, \dots$$

At each time step we compute the proportion of vector entries that are within $\pm 5\%$ of the current equilibrium value, and the mixing time (when 95% of the boxes are covered) is noted, as described in Section 2.2.3. The results are shown in Figure 10b.

The region between the bottom and center compartments (spot b) has the lowest mixing time (83 rotations). The other spots exhibit much higher mixing times: the top spot (a) has a mixing time more than twice as long (172 rotations), while the spot in the bottom compartment (spot c) has the highest mixing time (209 rotations). The spot (d), chosen for comparison with the experiments yields a mixing time of 201 rotations and therefore corresponds almost exactly to the one reported in Fitschen⁵³ of 195 rotations for a rotation rate of 249 rpm. The results for the mixing times using the experimental trajectory data within our transfer operator approach are also in the same range (see Figure S7 in the Supporting Information). A comparison with the mixing times retrieved directly from LB simulations, see Figures S8 and S9 in the Supporting Information, shows qualitatively a similar picture with the mixing times being 92 stirrer rotations for spot (b), 130 rotations for spot (a), 162 stirrer rotations for spot (c), and 173 stirrer rotations for spot (d) for the mean over ten runs. The lowest and highest mixing time over all ten runs are 61 stirrer rotations, corresponding to spot (a), and 186 stirrer rotations, corresponding to spot (d).

This computational mixing time analysis within the transfer operator framework adds a drastically improved method to perform detailed and simultaneously fast mixing time analysis studies in chemical reactors. Once the transition probabilities are calculated on Lagrangian data, any initial concentration distribution can be analyzed with minor computational effort. Here, the evolution of a density (e.g., a vector of

concentrations) via sparse matrix-vector multiplications and comparison with the stationary distribution for 250 rotations or time steps (as in Figure 10b) is computed in less than 2 s on a laptop.

5. CONCLUSION

The present study is the first application of classical transfer operator-based computational methods to analyze transport and mixing in a stirred tank reactor. The transfer operator is approximated by a stochastic matrix, where we have estimated the transition probabilities directly from given simulated or experimentally measured Lagrangian trajectory data. Former work has derived compartments for CM models exclusively via non transient, mostly averaged or single snapshot velocity fields,^{3,9,51} even though, this frozen flow field hypothesis limits the ability of the CM models to capture dynamic changes in the flow. The method we propose closes this gap in model design and is developed using the full transient Lagrangian dynamics of the fluid.

The transfer operator method itself can be interpreted as a compartment model with the small boxes from the discretization of the domain forming fully mixed compartments. The final Markov state modeling using Lagrangian coherent flow structures as compartments is an even more coarse-grained CM and reveals the macroscopic transport dynamics. We have motivated the use of almost-invariant or finite-time coherent sets as such coarse-grained compartments, as they could be highly mixing inside but are characterized by little material transport between them. We find very similar, spatially nearly fixed Lagrangian compartments in experimental and simulated STR data. These results are insensitive to the numerical parameters (see Figures S1, S2 and S6 in the Supporting Information) and the resolution of the trajectory data (Figure S4).

Also, the method proposed naturally determines the optimal number and shape of compartments to capture the transient Lagrangian dynamics via the eigengap analysis. This is an advantage in contrast to other methods that try to determine or automatize the optimal set and number of compartments in CFD derived CM models.^{9,51} For this work, the focus was set on the development of the Lagrangian transfer operator-based method. In the future, however, the methods proposed here can also be directly compared to other methods of compartment modeling since from our considered experimental and numerical data also average velocity fields can be calculated. These thus form a basis for Eulerian based compartment model derivation.^{3,8} The analysis of the internal mixing inside each compartment could further be used to decide with which reactor type the compartment may be modeled in CM, perfectly mixed or completely segregated.

Further, the transition matrix has been used to very efficiently compute expected residence times and mixing times for different initial concentration distributions mimicking different substrate feed locations. The proposed method is advantageous over methods that need to do elaborate computations of the concentration evolution for each feed location.^{9,51} Equally, the transition matrix based mixing time and residence time calculation can be used to compare different flow regions and different stirring protocols. We note that the transfer operator approximation in terms of a stochastic matrix induces numerical diffusion of the size of boxes in the discretization of the domain. Moreover, we have included additional diffusion to regularize numerical artifacts

resulting from the nonuniform distribution of tracer trajectories. How this can be related to the effective diffusion of the underlying system is subject to future research. The mixing times obtained within our framework can be seen as computationally very efficient approximations that are robust with respect to numerical parameters (see Figure S3 in the Supporting Information) and data resolution (Figure S5). They can be interpreted as mean quantities that compare reasonably well with those found in experiments and Lattice Boltzmann simulations (see Figures S8 and S9). We note that mixing times fluctuate significantly over different experimental or simulative runs. Our proposed approach can give a good indication of the mixing properties of a stirred tank reactor using very limited Lagrangian information (see also Figure S10).

One limitation of the classical transfer operator method is that a large number of trajectories that are well distributed in space is required to estimate the transition probabilities to sufficient accuracy. By inducing additional diffusion and cropping trajectories, when there is a certain time-periodicity, we have enriched the trajectory data in the current system and were able to capture the dynamics of the mass transfer.

Modern reactors have more complex geometries and dynamics compared to the classical stirred tank reactor, and the simple assumption of periodic dynamics is no longer possible. In this case, the compartments can be computed by applying the finite-time coherent sets framework. However, for sparse data network-based methods for the detection of coherent flow structures from trajectories as already used in a previous STR simulation setting⁴¹ might be more convenient. Moreover, adaptations of this approach can be used to track coherent sets over long time spans⁴⁰ and to evolve scalar fields in a purely data-based manner.⁵⁴ Current work includes extending these methods to the case of experimentally measured trajectories (see Steuwe et al.¹³ for a first study).

To optimize chemical reactors a better understanding of the fluid dynamical processes determining transport and mixing is crucial as the local reaction rates depend heavily on the local concentrations of the reactants and our present study is meant to provide a first step in that direction. It delivers a valuable tool to efficiently analyze the aforementioned phenomena and, when coupled with chemical kinetics, to approximate the outcome of a chemical reaction. With the developed methods, different substrate feed positions and reaction pathways can be tested extremely fast using a very simplified but meaningful CM model that bases on Lagrangian transport and therefore captures the true system dynamics. To incorporate chemical reactions we extend the method proposed here, where we fuse the evolution of density vectors by numerical transfer operators with update schemes involving chemical reaction kinetics.⁵⁵ We also plan to extend these ideas to the trajectory-based setting.

Although the methods presented in this work are exclusively applied to an STR, they are not restricted to this kind of apparatus and can be expanded to different types such as tubular reactors equipped with lattice structures and flows close to active boundaries. Furthermore, also scale-down models of industrial reactors that are of interest in bioreactors⁵⁶ can readily be studied with regard to concentration heterogeneities and mixing times with the proposed methods.

Finally, we aim to develop neural network approaches in order to analyze and quantify transport and mixing from very

sparse trajectory data, for instance provided by Lagrangian sensors in industry-scale reactors. The ability to assess the internal state of the reactor in real time (e.g., with respect to mixing, dead zones etc.) would be a crucial piece in the development of SMART reactors in the course of the Collaborative Research Centre 1615.

■ APPENDIX A

A. Set-Oriented Algorithms for Almost-Invariant and Finite-Time Coherent Sets

The following algorithm A.1 for finding almost-invariant sets has been adapted from Froyland and Padberg-Gehle,⁴³ whereas algorithm A.2 for approximating finite-time coherent pairs on M has been adapted from Froyland et al.⁴⁴

A.1. Set-oriented approximation of almost-invariant sets

- (1) Partition the state space M into a collection of connected sets $\{B_1, \dots, B_n\}$ of equal size.
- (2) Construct the Ulam matrix P using (6) (or the data-based versions (7), (8)), and compute the (assumed unique) fixed left eigenvector p of P .
- (3) Construct the matrix $\hat{P}$ where $\hat{P}_{ij} = \frac{p_j P_{ji}}{p_i}$ and $R = (P + \hat{P})/2$.
- (4) Compute the K largest nontrivial eigenvalues $1 > \lambda_2 \geq \dots \geq \lambda_K$ of R and corresponding eigenvectors $v_2, \dots, v_K$.
- (5) Extract almost-invariant sets from $v_2, \dots, v_K$.

A.2. Set-oriented approximation of finite-time coherent sets

- (1) Partition the domain M into a collection of connected sets $\{B_1, \dots, B_n\}$ of equal size.
- (2) Set $p = \frac{1}{n}\mathbf{1}$, where $\mathbf{1}$ is the all-ones vector as the volume measure to be tracked.
- (3) Construct the Ulam matrix P using (6) (or the data-based versions (7), (8)), and compute $q = pP$.
- (4) Define diagonal matrices $(D_p)_{ii} = p_i$ and $(D_q)_{ii} = q_i$, $i = 1, \dots, n$, compute the K largest nontrivial singular values $1 > \sigma_2 \geq \dots \geq \sigma_K$ of $D_p^{1/2} P D_q^{-1/2}$ and corresponding left and right singular vectors $\tilde{v}_j, \tilde{w}_j$, $j = 1, \dots, K$. Set $v_j := \tilde{v}_j D_p^{-1/2}$, $w_j := \tilde{w}_j D_q^{-1/2}$.
- (5) Extract finite-time coherent sets from $\{v_2, \dots, v_K\}$ and $\{w_2, \dots, w_K\}$.

Steps 5 in both algorithms address the extraction of sets from the leading eigenvectors. We use SEBA⁴⁷ for this as outlined in Section 2.2 in order to obtain K almost-invariant or finite-time coherent sets plus the remainder of the domain as the $(K + 1)$ -th set.

■ ASSOCIATED CONTENT

Data Availability Statement

The simulated STR data are available at [10.15480/882.16844](https://doi.org/10.15480/882.16844),⁵⁷ the experimental STR data are available at [10.15480/882.17469](https://doi.org/10.15480/882.17469).⁵⁸ The MATLAB code used for the numerical studies is available at [10.5281/zenodo.21874119](https://doi.org/10.5281/zenodo.21874119).⁵⁹ The code requires the MATLAB-toolbox for the GAIO library⁴⁶ (available at <https://github.com/gaioguy/GAIO>).

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.iecr.6c01170>.

It includes detailed computational analyses of the influence of numerical parameters on the resulting dynamical compartments and mixing statistics as well as additional results for the experimental data. Moreover, we present a validation of the mixing times by Lattice Boltzmann simulations (PDF)

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Notes

The authors declare no competing financial interest.

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